Modeling the "Shrink-Wrap Effect" in Polymers and Elastomers, Including the Influence of Very Large Elastic and Inelastic Strains

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Abstract: Thermally activated irreversible shrinkage of polymers and elastomers is an important characteristic that is often critical to the functional behavior of structures made from these materials. Shrink-wrap packaging is a common example of this phenomenon. When exposed to sufficiently hot air, the polymer film irreversibly shrinks significantly to create a taut enclosure. In some elastomers the results can be astounding. For example, an elastomeric specimen is stretched to 400% nominal strain. Then upon completely releasing the external load, the specimen still has 200% nominal strain remaining. However, after placing the specimen into boiling water, it recovers back to within 5% of its original shape. Modeling such material behavior is extremely challenging because the material law must accommodate large elastic and large inelastic strains as well as a mechanism to cause such large irreversible shrinkage. This paper presents intriguing material data for a thermoplastic elastomeric copolymer and discusses initial investigations of modeling such behavior using existing ABAQUS material subroutines.

1. Introduction

Certain classes of polymers and elastomers exhibit an irreversible shrinkage when they are exposed to sufficiently elevated temperatures. Thin polymeric films used for shrink-wrap packaging and shrink-wrap insulation on windows are common examples. Heating these films with hot air will cause them to permanently shrink. Upon cooling back to room temperature, the shrinkage remains. The amount of shrinkage depends on the film's material properties and previous thermal/mechanical loading history. This so-called "shrink-wrap effect" can be very pronounced and is often critical to the functional behavior of structures made from such materials. In the case of shrink-wrap packaging, too much shrinkage can cause excessive loads on the underlying package being wrapped; leading to bent corners on cardboard backers for example. If the shrinkage is insufficient, then the inner components of the package may move undesirably.

Another example of the shrink-wrap effect comes from a child's toy called "The Incredible Shrinky Dink Machine". In this case, heating from the Shrinky Dink machine causes a thick polymeric film to irreversibly shrink to 1/3 of its original planar length dimensions (*x* and *y*) while obtaining a 9 times increase in its original thickness (*z*). These dimensional changes remain upon cooling back to room temperature. This example clearly demonstrates the *apparently-orthotropic* shrinkage that typically occurs. The film had a positive shrinkage coefficient in the plane but a negative shrinkage coefficient through its thickness. Note, however, the Shrinky Dink film deformation was volume conserving. Similar volume conserving behavior also occurs generally with the thin shrink-wrap films, although the thickness change is often not reported.

Thermoplastic elastomeric copolymers have demonstrated astounding shrinkage behavior. These "elastomers" can endure very large amounts of deformation (greater than 800% nominal strain) without complete failure. However, when they are unloaded, significant permanent deformations remain. For example, a specimen is uniaxially stretched 400% Biot (nominal) strain. Then upon completely releasing the external load, the specimen has 200% permanent Biot strain remaining. Next, the specimen is placed in boiling water and the specimen shrinks significantly. The specimen is then removed from the water and returned to room temperature (with negligible additional changes in dimensions). After this "boil-off" treatment, the specimen has recovered back to within 5% of its original shape! In this example, the shrinkage in the axial dimension was positive (it became shorter) while the cross-sectional dimension become larger (exhibited a negative shrinkage). As was the case with the Shrinky Dink film, all of the deformation in the thermoplastic elastomeric copolymer was volume conserving.

These last two examples indicate clearly that this thermally activated shrinkage is not like the common CTE (Coefficient of Thermal Expansion) effect, but more like an annealing of inelastic stresses (where an inelastic deformation is volume conserving). Thus, what might appear at first glance to be a highly orthotropic material behavior, is likely not. As I will show shortly, this *apparently-orthotropic* shrinkage behavior can be explained with isotropic mechanisms.

All of the previous examples exhibit the following behaviors:

1. Thermally activated, *apparently-orthotropic,* irreversible shrinkage with apparent shrinkage coefficients that are positive and negative.

2. Constant volume deformations.

3. Large strains.

Modeling such material behavior is extremely challenging because the material law must have a mechanism to describe the shrinkage as well as accommodate large strains.[1] In the case of thermoplastic elastomeric copolymers, both the elastic and inelastic strains may be very large. The remainder of this paper discusses measured material data for a thermoplastic elastomeric copolymer and various approaches to simulate such behavior using existing ABAQUS material subroutines.

1.1 Measures of Strain and Stress

This section provides a brief overview of various measures of deformation, strain, and stress that will be used throughout this paper. Because we will be dealing with very large deformations, it is important to clearly define what measures are being used. For simplicity, we will define most of the quantities based on uniaxial (1-D) or principal measures; expansion of these concepts to tensors follows commonly used methods.

Deformation is most generally defined using the concept of the stretch ratio λ,

$$\lambda = L_c/L_{ref} \tag{1}$$

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1. It is important to note that not all materials will exhibit the shrink-wrap effect. For example, metals and "pure" elastomeric materials do not exhibit the effect, in general.

where L_c is the current length and L_{ref} is the reference length. It is generally desirable that the reference length is defined relative to an original (virgin) state before any deformation has been applied. However, this need not always be the case. With many polymers and elastomers (and other materials, including metals), the original (virgin) state is commonly unknown because the material has experienced significant inelastic deformation (via manufacturing processes) prior to ones receipt of said material for evaluation. For the shrinkage data that is presented shortly, this is indeed the case. Moreover, properly characterizing the reference state is one of the key factors to describing the shrink-wrap effect in a general manner.

Various measures of strain can be defined from the stretch ratio. We will use two forms. The Biot or nominal strain

$$\varepsilon^{Biot} = \lambda - 1 = (L_c/L_{ref}) - 1 \tag{2}$$

and the logarithmic or true strain

$$\varepsilon^{ln} = \ln(\lambda). \tag{3}$$

Two commonly used stress measures are the nominal stress and the Cauchy stress. The nominal stress is simply

$$T = F/A_{ref} \tag{4}$$

where F is the current force and A_{ref} is the reference area (which does not change or get updated during the deformation). The Cauchy stress is

$$\sigma = F/A_c \tag{5}$$

where A_c is the current area (which can change with the deformation).

It is important to note that the Cauchy stress is different from the nominal stress in that its value is independent of the reference configuration (it is always based on the current deformed state). In the analyses that follow, it will be seen that the logarithmic strain and Cauchy stress have certain computational and analytical advantages to processing data. However, it will also be seen that both the Cauchy stress and logarithmic strain have a compression effect on large-strain data; making it difficult to visually see certain patterns in the data. Using the Biot strain and nominal stress in these cases allows for improved data interpretation. What is most important to remember is that we can map back and forth between the different measures (and even different reference configurations) provided that we clearly keep track using equations of large deformation mechanics (as will be shown shortly).

2. Phenomenological description the shrink-wrap effect

The mechanisms which cause the shrink-wrap effect are fundamentally different from those which cause the more traditional behavior of thermal strains (expansion or contraction). In thermal expansion, increases in temperature cause the kinetic energy of molecules to increase which in turn causes a specimen's volume to increase. This process can be completely reversible; reduction of the temperature back to its original value will result in a specimen's volume returning to its original value. The thermal strains only act on the dilatational (volumetric) portion of the strain tensor. In general, we would expect that the isotropy of the thermal strain behavior is likely to follow the isotropy of other mechanical characteristics for a given

material. For example, it would be highly unlikely that a material which is mechanically isotropic to exhibit highly orthotropic thermal expansion (especially with both positive and negative coefficients).

In contrast, the primary mechanism causing the shrink-wrap effect is well explained with a two-segment annealing model. It is useful to idealize the material as being composed of two types of components (existing in parallel, i.e. they experience the same total strain): one that exhibits mostly hyperelastic deformation and one that deforms mostly inelastically. Further imagine that in the material's virgin state, there are no residual strains or stresses. If we took a sample of this virgin material (Figure 1, config: o) and stretched it sufficiently to produce significant inelastic deformation of the inelastic segment (Figure 1, config: a), then upon release of the external constraints, the sample would have a large permanent deformation remaining (Figure 1, config: b). In this released state, the hyperelastic segment would be in tension and the inelastic segment in compression. Additionally, the axial strain would be positive while the cross-direction strains would be negative (as measured relative to config: o). If the specimen was then heated such that the inelastic segment annealed, it would no longer be able to support load and the hyperelastic segment would be free to retract back to a zero stress, unstrained state (Figure 1, config: o). The inelastic segment freely deforms back along with the hyperelastic segment. Relative to the stressed state (Figure 1, config: b), the material looks like it had a very large, apparently orthotropic, shrinkage caused by thermal loading only (the length shrunk and the cross-section became bigger). Note that the deformation throughout this entire history would be essentially volume conserving because both the inelastic segment and the hyperelastic segment deformations are volume conserving.[1] In summary, the two-segment annealing model, using only isotropic material properties, represents the physically observed behaviors of the shrink-wrap effect, include *apparently*-orthotropic shrinkage. The key is that prior to thermal loading, the material has had sufficient deformation to induce inelastic compressive strains into the inelastic segment of the material and that the hyperelastic segment is left in a tensile state.

3. Material testing results for a thermoplastic elastomeric copolymer

Several tests were performed on a thermoplastic elastomeric copolymer to further investigate their shrink-wrap behavior. The specific material investigated was a poly(ether-ester-amide) elastomeric block copolymer composed of a Nylon 12 hard block (25% by weight) and a poly(THF) soft block (75% by weight). For simplicity, we will refer to this material as a "PEEA elastomeric copolymer". See McLean, 1999, for further descriptions of such materials. This material was chosen for study because of its unique properties and its ability to exhibit very large elastic and inelastic strains.

3.1 Boil-off shrinkage

Boil-off shrinkage is an easy method for inducing thermal shrinkage into an elastomeric sample. Both the elevated temperature and increased humidity act to anneal the inelastic segments in the material. The boil-off shrinkage procedure for uniaxial specimens is performed as follows:

1. The length of the specimen prior to shrinkage is measured at room temperature and denoted as L_{before}

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1. Elastically recoverable volumetric thermal strains are also produced but they are negligibly small compared to the other strains in the material.

2. The specimen is boiled, dried, and returned to room temperature. The length after boil-off is measured and denoted as L_{abo} (where "abo" denotes after boil-off).

The shrinkage of the sample is then computed as

$$\text{shrinkage} = 1 - \frac{L_{abo}}{L_{before}} \tag{6}$$

Comparing this with Equation 2, we see that shrinkage is just a negative measure of Biot strain with the length *before* shrinkage taken as the reference length.

Table 1a presents shrinkage results for three different versions of the PEEA elastomeric copolymer. All three material versions have identical material make-up, only their prior deformation history is different. The 1X material is a virgin sample (never been stretched). The 2X and 4X versions have been stretched during their manufacturing to stretch ratios of 2.0 (same as 100% Biot strain) and 4.0 (same as 300% Biot strain), respectively, and then released (creating residual stresses and strains). The data from Table 1a indicates that version 1X exhibited little shrinkage while versions 2X and 4X exhibited large amounts of shrinkage. Based on the phenomenological description of the shrink-wrap effect (Section 2), we hypothesize that if no other constraints are applied to the specimen during boil-off, then after boil-off the material should return to its original unstressed state (as it was in its virgin state). This implies that the length *after* boil-off should be the reference state for measuring deformation and that the material specimens had initial strains *before* boil-off (as we know was the case). Thus, we estimate the initial strain *prior* to boil-off as

$$\varepsilon_{pbo}^{Biot} = \frac{L_{before}}{L_{abo}} - 1 = \frac{\text{shrinkage}}{1 - \text{shrinkage}} \tag{7}$$

These results are also reported in Table 1a.

If these hypotheses are true, then further stretching of the materials after boil-off should yield similar results for all three material versions. To verify this, 200% Biot strain (relative to their lengths after boil-off, L_{abo}) was applied to all three specimens. The applied strain was then released and the remaining permanent set in each specimen was measured. The specimens were then re-boiled and their final strains were measured. These results are reported in Table 1b. Note that all these strain measures use the Biot strain formula from Equation 2 where the reference length is L_{abo} (the length *after* the original boil-off) and the current length is the specimen length after the given testing action. For the strain ranges tested, all three material versions tested nearly identical and thus supports the residual stress hypothesis.

Looking closer at this data, we see that boiling did have an additional effect. The samples that were stretched prior to any boil-off (during their manufacturing, samples 2X and 4X), had permanent sets of approximately one half of the applied manufacturing strains (64% initial strain relative to 100% applied strain and 150% initial strain relative to 300% applied strain). However, stretching the material 200% after it was boiled-off produced a permanent strain of only 20%, which is 10% of the applied strain. Note that the 2X and 4X material was stretched during its manufacturing process when it was not fully cured. We would expect the residual strains from this to be somewhat different than from a cold draw at room temperature of fully cured material (as was the case for strains applied after boil-off). As a result, it is not clear how to exactly interpret the differences. What is clear, though, is that boiling the material did release significant residual strains. It is also noted that all the specimens for these tests showed negligible volume changes.

3.2   Monotonic, uniaxial loading

Monotonic loading of long, round, uniaxial samples using an Instron was performed on the three different versions of specimens, namely 1X, 2X, and 4X. As stated earlier, versions 2X and 4X had been inelastically deformed during manufacturing. However, none of the samples were boiled for this portion of the evaluation.

Load and cross-head displacement[1] was measured and then converted to nominal stress and nominal strain. Figure 2a presents the resulting nominal stress vs. nominal strain data for each of the three samples using a common method of data processing that most material test labs would utilize. This method is summarized as follows:

1. Prior to testing, record the specimen gage length L_s and cross-section area A_s (subscript "s" stands for starting state).

2. During the test, record the current length of the specimen L_c as a function of the applied load F.

3. Compute the stretch ratio, nominal strain, and nominal stress all relative to the starting state as

$$\lambda_s = L_c/L_s \tag{8}$$

$$\varepsilon_s^{Biot} = \lambda_s - 1 \tag{9}$$

$$T_s = F/A_s \tag{10}$$

The resulting stress-strain plots shown in Figure 2a seem to indicate that the three different specimens have very different material responses (they appear to act as though they are different materials). However, we know that they are identical materials, and that only their deformation histories prior to testing are different. The crux of the problem is in our choice of reference configuration. We need to evaluate the data based on the specimens original, virgin state (prior to any deformation history). The equations that follow will allow us to map the nominal stress and Biot strain data computed relative to the starting state to any other state we need.

We first define the following stretch ratio measures:

$$\lambda_{init} = L_s/L_o \tag{11}$$

$$\lambda_o = L_c/L_o = \lambda_{init}\,\lambda_s \tag{12}$$

where λ_{init} is the initial stretch ratio of the specimen at the beginning of the test and λ_o is the true stretch ratio based on the original (virgin) length of the specimen L_o. The initial stretch ratio characterizes the initial strain in the specimen at the starting state of the test. It is noted that none of the quantities describing the initial stretch, original length, or original strain are known prior to the test. The methodology that follows shortly will describe how we estimate them from a master curve.

Corresponding areas for the current and original states are denoted as A_c and A_o, respectively. The volume ratios and area ratios then follow via

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1. Because the specimens were very long ($\gg$ 10x1 aspect ratio) and the deformations are very large, cross-head displacement can be used to accurately measure sample elongation.

$$J_s = \lambda_s \frac{A_c}{A_s} , \quad J_o = \lambda_o \frac{A_c}{A_o} , \quad J_{init} = \lambda_{init} \frac{A_s}{A_o} \tag{13}$$

Note that for many materials, the deformation is essentially volume conserving and all the volume ratios can be assumed to equal 1.0.

Biot and logarithmic (true) measures of strain relative to the various stretch ratios are defined as

$$\varepsilon_s^{Biot} = \lambda_s - 1 , \quad \varepsilon_o^{Biot} = \lambda_o - 1 , \quad \varepsilon_{init}^{Biot} = \lambda_{init} - 1 \tag{14}$$

$$\varepsilon_s^{ln} = \ln(\lambda_s) , \quad \varepsilon_o^{ln} = \ln(\lambda_o) , \quad \varepsilon_{init}^{ln} = \ln(\lambda_{init}) \tag{15}$$

Noting the law of logarithms we find that the true strain relative to the original state is simply the sum of the true strain measure of the initial strain plus the true strain measure relative to the starting state

$$\varepsilon_o^{ln} = \varepsilon_{init}^{ln} + \varepsilon_s^{ln} . \tag{16}$$

Equation 16 demonstrates that the difference in strain measures between the different reference states is simply a shift caused by the initial strain. This fact is only true when measuring strain with the logarithmic measure, it is not true when using the Biot measure of strain (nor any other measure such a Green or Almansi strains).

The nominal measure of stress relative to a cross-sectional area based on the starting state was defined in Equation 10. The nominal stress relative to the original state is similarly computed as

$$T_o = \frac{F}{A_o} = \frac{J_{init}}{\lambda_{init}} T_s \tag{17}$$

The Cauchy (true) stress, Equation 5, is always defined as the force relative to the current area, regardless of the reference state. Substituting Equations 10 and 13 into Equation 5 allows us to convert the nominal stress relative to the starting state to the Cauchy stress as

$$\sigma = \frac{F}{A_c} = \frac{\lambda_s}{J_s} T_s \tag{18}$$

These previous equations allow us to readily map strain and stress results between different measures and different reference configurations. Unfortunately, we do not know a priori what the original (virgin) state of the specimens are nor the initial strain in the specimens. One approach to estimate this is to use the boil-off shrinkage data of Section 3.1. This was attempted by the author, but the results were less accurate for large initial strains (especially for the 4X samples).[1]

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1. Detailed analysis of the boil-off data and the stress-strain data indicates that some permanent damage occurs at very large strains, say above 200% Biot. The boil-off hypothesis used to estimate initial strains assumes that all the inelastic strains are recoverable upon boil-off. If the material has significant permanent damage, then this hypothesis would produce errors.

An alternate approach using a curve-fit technique to determine the initial strain that existed in the material is used instead of the boil-off approach. This method requires a measured stress-strain data set of a virgin material (no initial strains or stresses). This virgin material response is termed the Master curve. For the PEEA elastomeric copolymer tested in this paper, the 1X specimens are used for the Master curve since they had negligible pre-stretching during manufacture and they showed negligibly small shrinkage from the boil-off tests. To find the initial strains in the 2X and 4X specimens, the following procedure was used:

1. Plot the Cauchy stress vs. Logarithmic strain for the virgin material (denote this as the Master curve). Note that for a virgin material, the initial stretch ratio is $\lambda_{init} = 1$.

2. For each specimen in question, convert the nominal stress relative to the starting state, T_s, to Cauchy stress, σ, via Equation 18. Additionally, convert the stretch ratio relative to the starting state, λ_s, to logarithmic strain relative to the starting state, ε_s^{ln} via Equation 15.

3. Overlay this specimen data onto the Master curve and simply shift the specimen data on the strain axis to determine a best fit. The amount of strain shift needed defines the amount of initial logarithmic strain that exists in the sample.

Figure 2b demonstrates the result of this analysis for the 2X and 4X specimens. The logarithmic measures of the initial strains for the 2X and 4X were determined to be 60% and 115%, respectively. These equate to initial stretch ratios of 1.822 and 3.158, respectively. Comparing these two initial stretch ratios to those from the boil-off analysis (Table 1a), we see that the values here indicate more initial strain than those determined from boil-off. If the boil-off does not remove/repair *all* the initial inelastic strains (which is likely true), then we would expect the boil-off estimate of initial strains to be lower than those computed from the stress-strain data.

Looking at the curves in Figure 2b, we also see another difficulty when dealing with very large strains; namely data compression. The measures of Cauchy stress and logarithmic strain tend to compact the data at large strains, making it difficult to interpret the curves easily. Using Equations 14, 15, and 17, we can convert the Cauchy Stress vs. Logarithmic strain back to nominal stress and Biot strain (both relative to the original, virgin, state). This is depicted in Figure 2c and tends to expand the data, showing details better. Note that it is valid to plot nominal stress-strain data because the data is now mapped to a common state for all samples, namely the original virgin state.[1]

3.3   Cyclic Loading

Figure 3 depicts the experimental data for the PEEA elastomeric copolymer when it is cyclically loaded. The same analysis approach used in Section 3.2 and Figure 2 is used here. To avoid clutter, only the final adjusted data, plotted in terms of nominal stress vs. Biot strain (both relative to the original, virgin state) is shown. The arrows on the plots denote the direction and cycle of the load path (1 - apply tensile load, 2 - release load, 3 - apply tension again, 4 - release load again). The data shows excellent correlation for the various specimens. This cyclic data also shows the significant amounts of permanent set that can occur in

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1. If one attempts to perform the curve-shift using nominal stress and nominal strain data (in the original configuration), the curve-fit analysis is more complicated because both stress and strain values are effected by the unknown initial strain. Additionally, the strain axis no longer simply shifts, but it also stretches as a function of initial strain.

the material. The important thing to remember about this material relative to the shrink-wrap effect is that upon sufficient heating, these huge permanent sets of between 25% and 400% Biot strain (Figure 3) essentially vanish as the material shrinks to recover nearly all this strain! (not explicitly shown in these figures).

The problem now is "given this material data presented in Section 3, how does one model it in ABAQUS?".

4. Initial investigation of modeling approaches in ABAQUS

The general approach to modelling such complex behavior is to characterize the material using the total deformation gradient, F, defined as

$$F = \frac{\partial x}{\partial X} \qquad (19)$$

where x is the position in the current configuration of a material particle that was at position X in the original configuration. Then, via a multiplicative decomposition of the deformation, we separate the deformation gradient into elastic and inelastic deformations as

$$F = F^{elas} \cdot F^{inelas} \qquad (20)$$

Unfortunately, continuing down this path leads to an extremely complicated analysis, a self written user material subroutine, ..., and a major research project!

So the question is asked, "what can we do with current features in ABAQUS?" Our initial investigations evaluated three possible approaches.

4.1 Effective CTE Approach

This approach uses artificial CTE constants to mimic the shrinkage strains observed in a given material. For simple cases with small shrinkage strains, this approach might be used. In general, you will need to use an orthotropic material law to specify the positive and negative shrinkage coefficients required to get correct "shrinkage" in all three directions (x, y, z). Having to choose orthotropic CTE values immediately eliminates any large strain hyperelastic formulations since ABAQUS only has isotropic hyperelasticity. Furthermore, the CTE approach does not allow one to study how initial deformation histories effect thermal shrinkage since it is not physically modeling the problem. These limitations plus several others suggest that the effective CTE approach is not well suited to model the shrink-wrap effect for general purpose applications.

4.2 *HYSTERESIS model

The experimental data presented suggests that the material has both hyperelastic characteristics plus inelastic characteristics. A material law that can combine both behaviors and then have the ability to anneal the inelastic portion of the material law might have a chance at representing the observed shrink-wrap effect. Utilizing *HYPERELASTIC and its suboption of *HYSTERESIS, we have a material model that may be able to mimic the needed material characteristics. The option as described in the ABAQUS/Standard User's Manual (Hibbitt, 2001) is designed to model strain-rate-dependent, hysteretic behavior of elastomers. While it is true that the material behavior of the PEEA elastomeric copolymer has strain-rate dependence, that is a secondary effect relative to the material data presented in Section 3. However, the general hysteresis form of the material law makes it appear to be a viable candidate. The basic concept will be to artificiality

manipulate the rate-dependent terms to produce behavior similar to the actual quasi-static experimental material data presented earlier. The *HYSTERESIS model uses a multiplicative decomposition of the total deformation gradient similar to that outlined in Equation 20. The essence of this material law is that the stress in the material is the sum of a purely hyperelastic component and a component that characterizes the time-dependent deviation from equilibrium. The two components are placed in parallel, i.e., they see the same strain. Note that this two-component hysteresis material law is very similar in many ways to the phenomenological description of the shrink-wrap effect given in Section 2.

The effective creep strain $\dot{\varepsilon}^{cr}$ of the time-dependent component is (Hibbitt, 2001)

$$\dot{\varepsilon}^{cr} = A(\lambda^{cr} - 1)^{C} \sigma^{m} \tag{21}$$

where A is a non-negative constant to maintain dimensional consistency, λ^{cr} is the creep stretch, C is restricted to the interval [-1, 0], and m is a positive exponent, generally greater than 1. In addition to these parameters, a stress scaling factor, S, is required to define the ratio of stress carried by the purely hyperelastic component and that carried by the creep component.

Figure 4b depicts the prediction of a uniaxial specimen cyclically loaded using the hysteresis approach. Values of the material constants shown were obtained via a trial-and-error approach. The general shape of the cyclic stress-strain curve looks reasonably similar to the experimental data. Unfortunately, initial evaluations with this model had only limited success because the hysteretic material law becomes numerically ill-conditioned when the creep stretch is near 1.0. Since the exponent C in Equation 21 is restricted to be a negative value, the creep strain rate equation blows-up when $\lambda^{cr} = 1$. The problem was discussed with HKS and at this time there is no apparent work-around for this issue. Initial tests with various cyclic loading profiles found that this numerical ill-conditioning caused repeated and unresolved numerical problems. As a result, this approach was abandoned until this issue can be resolved.

An additional comment related to this hysteresis approach is important. The hysteresis law, as implemented, has no provision to turn-off (or anneal) the creep strain and resulting stresses. Without such provision, we can only simulate how the residual stresses and strains are produced, but we can not evaluate the effect of their annealing (at elevated temperature). A potential work-around to this problem would be to use overlapping elements in the mesh. With this technique, the mesh has two identical elements where there would normally be one. Since the overlapping elements share the same nodes, each pair of elements will see the identical strain field and their stresses will be additive. This is identical to the basic approach of *HYSTERESIS, only now we effectively have a second stiffness in parallel. Using this approach in conjunction with *MODEL CHANGE would allow a method to simulate annealing of the inelastic portion of the material behavior.

4.3   Combining Hyperelastic with Elastic/Plastic via an overlapping element approach

This last technique evaluated combining *HYPERELASTIC with *ELASTIC and *PLASTIC using an overlapping element approach similar to the one described in the previous section. Instead of using *MODEL CHANGE to remove the inelastic elements to simulate the annealing which causes shrinkage, the new *ANNEAL TEMPERATURE option was used. In this model, for each pair of duplicate elements, one element has a *HYPERELASTIC material definition and the other element has a *ELASTIC and *PLASTIC material law definition. Because of the very large strains that need to be simulated, special care had to be taken when selecting the *ELASTIC and *PLASTIC material parameters to ensure that large elastic strains in the elements with *ELASTIC did not occur (*ELASTIC is designed to only accommodate

small elastic strains. Large elastic strains, say over 30%, can create significant errors and numerical problems).

Figure 4c demonstrates the prediction of an uniaxial specimen cyclically loaded with this modeling approach. Material parameters were obtained via a trial-and-error method. The results are not as good as the HYSTERESIS model, but they do produce some of the same basic features as the real data.

A simulation of thermal shrinkage for this model was presented in Figure 1. The stress-strain curves and deformed element shapes represent the individual material-segment stresses and the total combined stresses from this model as the material is initially stretched, unloaded, and then annealed with *ANNEAL TEMPERATURE. Using this approach, the model was able to simulate large amounts of shrinkage. This approach allows for annealing, in a controlled approach, of the inelastic stresses to simulate a thermal shrinkage. The primary deficiency of this model is that its ability to accurately simulate the generation of the residual stresses is only fair at best (as indicated in Figure 4).

One additional numerical issue occurred with this overlapping element approach. The models commonly produced negative eigenvalue warnings when the total applied nominal strains increased above 30% (and sometimes higher). Checks were made to insure that this was not caused by large elastic strains in the elements with *Elastic (these elastic strains were well under 5% for this strain range and never exceeded 10% for total applied Biot strains up to 200%). An initial analysis indicates that the double-element models preferred to twist at higher strains as opposed to simple uniaxial deformation. Further investigation is needed to better understand this issue.

5. Conclusions

Thermally activated irreversible shrinkage (the shrink-wrap effect) in polymers and elastomers can be an important characteristic that is often critical to the functional behavior of structures made from these materials. This paper has presented intriguing material data for a PEEA elastomeric copolymer under high strain deformations. Methods for properly processing initially stressed hyperelastic/inelastic material data have been demonstrated. Evaluating this and other material data has indicated that the large thermal shrinkages observed are not likely to be caused by traditional volumetric thermal expansion, but rather one of stress annealing from a pre-stressed, two-component material. Initial investigations of potential methods for modeling this behavior with existing ABAQUS material subroutines has also been presented. These results indicate that only moderate success can be achieved with the current material subroutines and that further development work would be needed for improved accuracy and generality.

References

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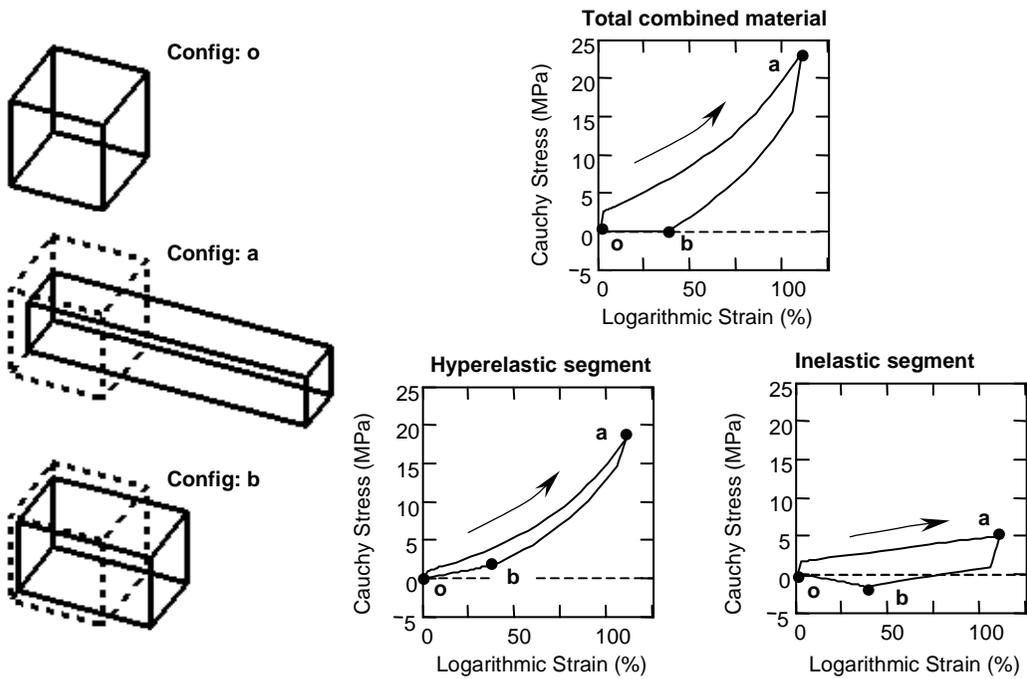
Table 1. Boil-off shrinkage of PEEA elastomeric copolymer

a.) Original boil-off results				b.) Results *after* initial boil-off			
	Specimen Versions				Specimen Versions		
	1X	2X	4X		1X	2X	4X
Shrinkage (%)	3	39	60	Applied strain $\varepsilon_{applied}^{Biot}$ (%)	200	200	200
Strain before boil-off, ε_{pbo}^{Biot} (%)	3	64	150	Permanent set ε_{set}^{Biot} (%)	21	20	20
Initial stretch ratio prior to boil-off, λ_{pbo}	1.03	1.64	2.50	Strain after re-boil $\varepsilon_{reboil}^{Biot}$ (%)	1	1	1

Total combined stress = stress from hyperelastic segment + stress from inelastic segment

Figure 1.   Phenominlogical description of the shrink-wrap effect.

a) Results relative to starting state configuration

Nominal stress, T_s (MPa)

150

100

50

0

0   200   400   600   800

Biot strain, ε_s^{Biot} (%)

4X

2X

1X

Figure (a) shows the data as would be commonly processed by a test lab. No accounting for initial strains is included.

b) Results relative to original (virgin) configuration

Cauchy stress, σ (MPa)

800

600

400

200

0

50   100   150   200

Logarithmic strain, ε_o^{ln} (%)

1X   2X   4X

c) Results relative to original (virgin) configuration

Nominal stress, T_o (MPa)

60

50

40

30

20

10

0

0   200   400   600   800

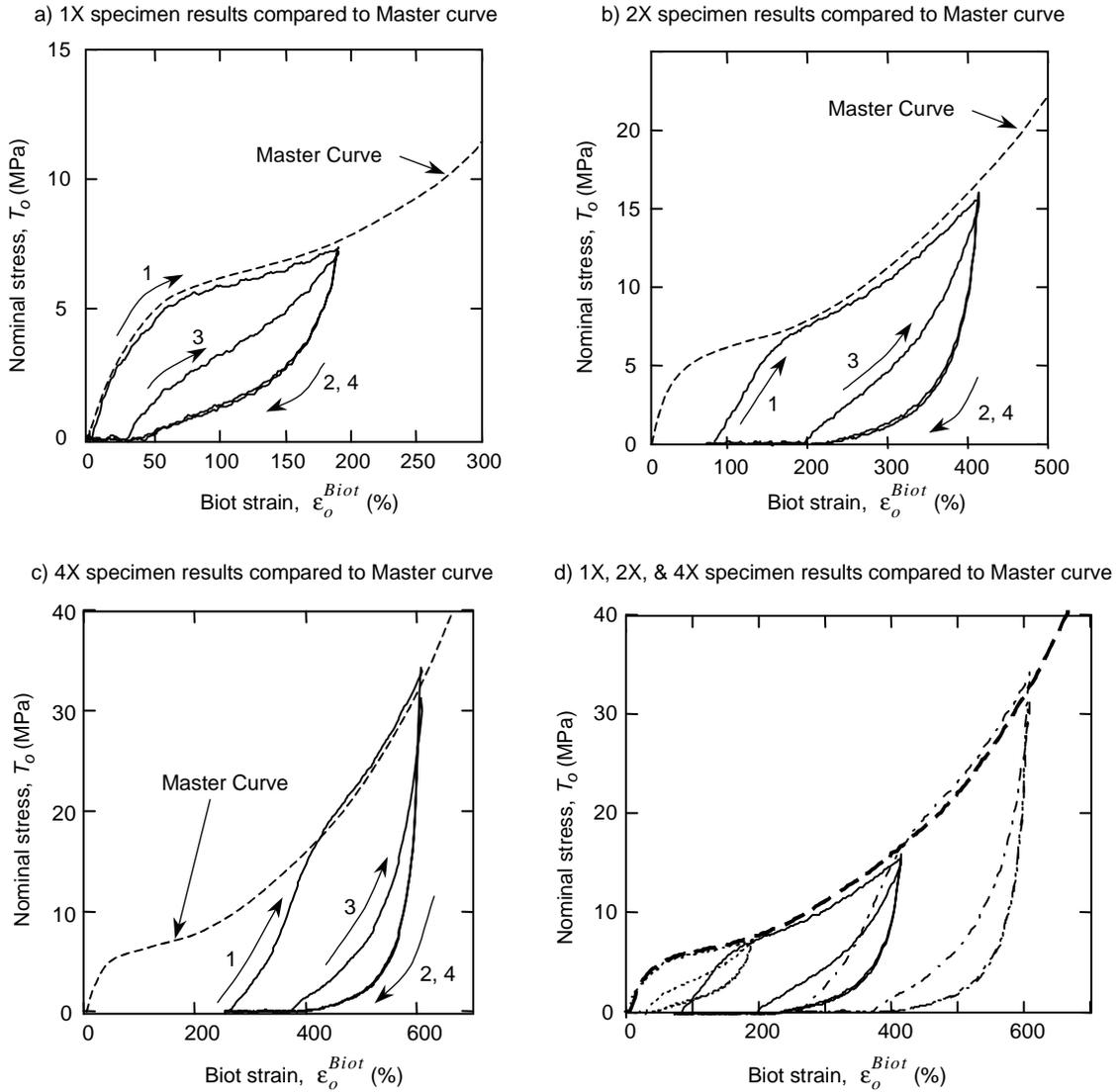Biot strain, ε_o^{Biot} (%)

1X   2X   4X

Note: For Figures b and c, the 2X and 4X curves have been shifted to account for initial strains. The best-fit shifts for initial strains for the various specimen types were:

1X: No initial strain shift, specimen assumed to be virgin (used as Master curve)

2X: $\varepsilon_{init}^{ln} = 60\%$

4X: $\varepsilon_{init}^{ln} = 115\%$

Figure 2.   Monotonic uniaxial stress-strain data for PEEA elastomeric copolymer.

a) 1X specimen results compared to Master curve

b) 2X specimen results compared to Master curve

c) 4X specimen results compared to Master curve

d) 1X, 2X, & 4X specimen results compared to Master curve

Note: For Figures a - d, the 2X and 4X curves have been shifted to account for initial strains. The best-fit shifts for initial strains for the various specimen types were:

1X: No initial strain shift, specimen assumed to be virgin (used as Master curve)

2X: $\varepsilon_{init}^{ln} = 60\%$

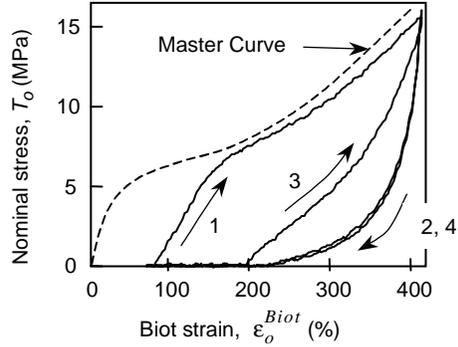4X: $\varepsilon_{init}^{ln} = 115\%$

Figure 3.   Cyclic uniaxial stress-strain data for PEEA elastomeric copolymer. Adjusted for initial strains & plotted relative to original (virgin) reference config.

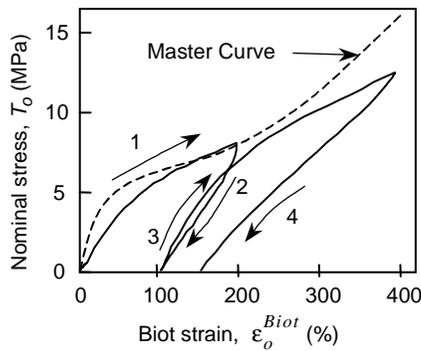**a) Experimental results from PEEA elastomeric copolymer**

1X specimen results compared to Master curve
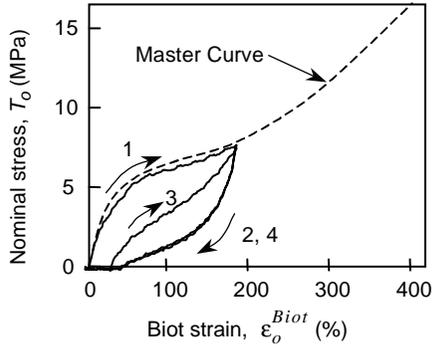
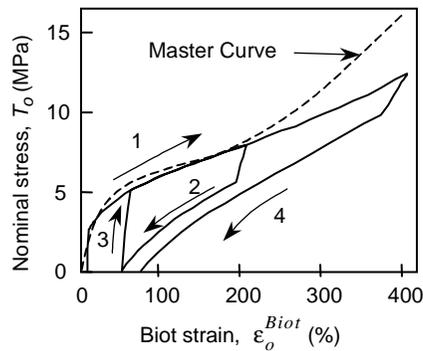2X specimen results compared to Master curve

b) FEA model: Hyperelastic with Hysteresis using single element approach


```
*MATERIAL, NAME=Hyper-Hyst
*HYPERELASTIC, ARRUDE-BOYCE
1.425, 5.688, 0.0
*HYSTERESIS
8.0, 0.028, 1.0, -1.0
```

c) FEA model: Hyperelastic with Elastic/Plastic using an overlapping element approach

Elset A - Hyperelastic component
```
*MATERIAL, NAME=Hyperelastic
*HYPERELASTIC, ARRUDE-BOYCE
1.710, 5.688, 0.0
```

Elset B - Elastic/Plastic component
```
*MATERIAL, NAME=Elastic-plastic
*ELASTIC
120.0, 0.4
*Plastic, Hardening=Combined,
Data Type = Parameters
2.4, 4.8, 0.2
*Cyclic Hardening, Parameters
2.4, 3.6, 0.2
```

Figure 4.   FEA results from two types of combined material models.