Elastic and Viscoelastic Recovery of Forged Polymer

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ABSTRACT

Although much research has been carried out in the past on solid-phase forming, little has been done on polymer forging. This can be partially attributed to the lack of fundamental understanding on instantaneous recovery and viscoelastic recovery of the forging, and consequently the lack of a modeling and simulation capability. In the present study, upsetting experiments were conducted to study the forging behavior of PTFE (poly tetrafluoroethylene). Both instantaneous elastic recovery and time-dependent post recovery were found to be significantly affected by the strain applied, the upsetting speed and the dwell time. Samples forged with higher forging speed had higher instantaneous recovery but lower time-dependent recovery. By employment of a dwell time, the instantaneous recovery can be effectively reduced. The dimensional recovery behavior of upset samples was qualitatively interpreted using the Vernar's model.

INTRODUCTION

Thermoplastic polymers are typically processed using various thermoplastic molding processes, e.g. injection molding, extrusion blow molding, compression molding, etc., in which the material is melted and shaped under pressure work [1]. These methods involve polymer fluidization by heating and shaping of the polymer inside the mold cavity, followed by cooling. In such processes the production rate is greatly affected by the cooling time because polymers in general have a poor thermal conductivity [2]. Other limitations of these processes arise from the difficulty in producing thick parts (the thickness usually limited to 3 mm) [2] and the difficulty in processing ultra high molecular weight polymers [1].

Polymers have low stiffness and strength in comparison with other materials and consequently have to be improved in properties prior to their applications. One effective way for enhancing the mechanical properties of polymers is to introduce molecular orientations to the polymer matrix. In particular, stiffness and impact strength can be significantly improved by the employment of molecular orientations [3]. Orientations can be induced in polymers either in the melt, rubbery or solid states [1, 3]. For example, when the polymer is deformed below its softening temperature, the deformation-induced orientation will be locked in the final part.

In the past half century a considerable amount of work [1, 3, 4] has been done to develop new processes different from the conventional molding process. These processes resemble more the solid-phase metalworking process, in which the work material does not have the fluidity to give rise to the type of fluid flow observed in processes such as extrusion and injection molding [4]. One advantage of polymer solid-phase forming is that the cooling time is considerably reduced. Further, solid-phase forming results in enhanced mechanical properties due to the self-reinforcing effect arising from molecular orientations. Polymer solid-phase forming has been successfully used in processing fibers, films, sheets and shell structures [3]. Examples of solid-phase formed polymer articles are cold drawn high-strength high-modulus polymer fibers, stretch blow molded bottles, biaxially orientated films, etc. In general, these techniques involve elongational deformation of a relatively thin cross section. Very recently, the technology was adapted to cold extrusion of polymer rods under extremely high pressures [5]. The so-extruded rod has superb mechanical properties due to a high degree of self reinforcing.

Another polymer solid-phase forming technique is forging [1, 4], which utilizes compression force to shape a relative thick material. Besides the general advantages associated with solid-phase forming, polymer forging is able to produce discrete parts, process difficult-to-mold polymers, and manufacture thick wall sections. For example, difficult-to-mold polymers such as ultra-high-molecular-weight polyethylene can be processed using this technique. Despite these advantages, polymer forging has been attracted much less attention than stretching or drawing based solid-phase forming processes. From the limited literature [1, 4] about polymer forging, it is conjectured that the hindered development of the polymer forging process could be partially attributed to the lack of understanding on the deformation and recovery behavior of solid polymers during forging. Some qualitative findings related to the polymer forging behavior can be drawn from the literature [1, 4]. It appears that springback remains the major problem. The severity of the springback problem is much more than for sheet forming processes because of the usual lack of symmetry and variation in section thickness in forging. The dimensional change of forged polymer occurs upon removal of the forging from the dies and during its storage at the room temperature. Such dimensional change varies with the change of materials, geometries and process conditions. With the lack of understanding on the dimensional

recovery process, it is hard to control the dimensional accuracy of forged polymer products. Further, such lack of understanding results in the lack of a suitable simulation capability for polymer forging.

From the fundamental point of view, the bulk deformation behavior of solid polymer during forging may be characterized using direct compression test or upsetting test. The related recent literature about deformation behavior of solid polymer under compression test is thus briefly reviewed in the following. Wang et al. [6] studied the deformation behavior of PP (polypropylene) under uniaxial compression. In their experiment, PP was subjected to compression at temperature between Tg (glass transition temperature) and Tm (melting temperature) and the deformation responses were studied by performing axisymmetric compression test. The friction was reduced by using a hot die and a proper lubricant. They found that the thermomechanical deformation of PP under compression was sensitive to temperature and strain rate. The stress-strain response was captured by an elastic yield at the early stage, a constant flow stress at the intermediate stage, and an orientation hardening at the final stage. Ray et al. [7] studied the stress-strain behavior of PE (polyethylene) and EVA (poly ethylene-co-vinyl acetate) blends under compression with various strain rates and temperatures. They found that stress-strain plots consist of three parts, namely, elastic or Hookean region, region of chain slippage and region of strain hardening and these regions are significantly affected by the change of strain rate and temperature. Pluta et al. [8] subjected polypropylene to plain strain compression and the deformed structure was investigated using light microscopy and differential scanning calorimeter. It was found that the initial sperulitic morphology was destroyed and was transformed into stacks of crystalline lamellae rotating towards loading direction. The deformed sample showed an increase in the melting point compared with the undeformed sample. The effect of friction on the deformation behavior of polymer during compression was also studied in the literature [6, 9, 10]. To reduce the amount of barreling a low coefficient of friction is often preferred. Lubricants can be used to reduce friction between the specimen and the platen [6]. Bruce et al. [9, 10] studied the molecular orientation of PP discs uniaxially compressed both lubricated and unlubricated. In the unlubricated sample the orientation was found to be inhomogeneous and was dependent on the radial distance from the disk axis. In the lubricated sample the orientation texture was perpendicular to the compression direction.

It can be seen that most of the previous work regarding the behavior of solid polymer during compression is about the flow stress data, i.e. stress as a function of strain. However, the flow stress data are not able to offer any insight into the dimensional recovery process after forging of the material. Therefore, systematic studies on the post compression behavior including elastic and viscoelastic recovery are needed in order to further the development of the polymer forging process. In the present study, upsetting experiments were conducted to study the deformation behavior of PTFE (poly tetrafluoroethylene), during compression and the dimensional recovery behavior after compression. The effects of different process variables including strain rate and dwell time on the room temperature forging behavior of PTFE were studied.

MATERIAL SELECTION CONSIDERATIONS

Similar to traditional metal forging processes, a high amount of ductility of the material is greatly desired in polymer forging. There are a wide variety of solid polymers available and the ductility of the material differs from one to another. Amorphous polymers with glass transition temperature (Tg) above the room temperature generally are brittle. The ductility of these polymers increases as temperature increases and the polymer becomes a rubbery material above its Tg. The ductility of semicrystalline polymers is also significantly dependent on the temperature that the polymer is subjected to. The role of the Tg on the ductility depends on the amount of crystallinity of the polymer. Generally speaking, a semicrystalline with a Tg well below room temperature is a highly ductile polymer. Polyethylene is an example, which is able to be elongated to more than 200% of its original length. In addition, the ductility of polymers also depends on the loading configuration. An amorphous polymer such as polystyrene, which is brittle under tensile test, could exhibit a ductile fracture under compression test.

A large amount of plasticity of the material is also highly desired in polymer forging. Although superplasticity can be achieved at elevated temperatures well above Tg for amorphous polymers and above melting temperature (Tm) for semicrystalline polymers, polymer solid is characterized by poor plasticity or high elasticity at low temperatures. Polymer solids thus exhibit a high amount of elastic recovery after forging, which is the major problem in polymer forging [4]. Therefore, selection of a material with a low elastic recovery is a priority. In general, glassy polymers show a high amount of elastic recovery compared to semicrystalline polymers and large deformation is possible only at temperatures close to Tg [4]. Hence, semicrystalline polymers are considered to be more suitable in polymer forging.

EXPERIMENTAL SETUP

PTFE, a semicrystalline polymer, was chosen as the materials in the study because PTFE has very good ductility and plasticity. PTFE is a difficult-to-mold polymer if melt processing is used because of its extremely high processing temperature and tendency of wall slip during molding, which results in a shark-skin type of rough surface of molded parts. The PTFE used in the study was 8735K virgin PTFE from McMaster-Carr Supply Company. The related material properties from the supplier are listed in Table 1. The material stocks are cylindrical bars with a diameter of 12.7 mm, which were produced using polymer extrusion.

Table 1: Room temperature mechanical properties of PTFE.

Tensile strength, MPa	*Elongation, %*	*Flex Modulus, MPa*	*Izod Impact Strength, J/m*	*Hardness, Shore D*
31	400	346-620	180	55

Upsetting experiments of 12.7-mm long PTFE cylindrical bars were carried out on a universal Instron testing machine. The sample was forged to a half of its original length. Three upsetting speeds were used, i.e. 12 μm/s, 48 μm/s and 144 μm/s. After unloading, the instantaneous elastic recovery and the time-dependent post recovery were measured and recorded. The effect of the dwell time was also studied. The dwell time is the time from the full stop of the load cell to the unloading instant. The dwell time used was 5 minutes. Each experiment with or without dwell time was repeated four times. The stress-strain response of the material was determined using the same upsetting setup.

RESULTS AND DISCUSSION

The true stress-strain curves for different speeds are shown in Figure 1. Strain and strain rate hardening effects can be seen in the figure. The elastic recovery modulus, i.e. the slope of the recovery curve, was found to be strain dependent. The elastic recovery modulus decreases as the strain increases. The instantaneous elastic recovery for a specific strain and upsetting speed can be directly read from the plot. For example, the elastically recovered strain for a compression strain of 1.38 (corresponding to a reduction of 75% of the sample height) and an upsetting speed of 144 μm/s is about 0.44. Therefore, about 30% of deformation was elastically recovered after unloading. Within the range of the strain tested, the material did not show a yielding point.

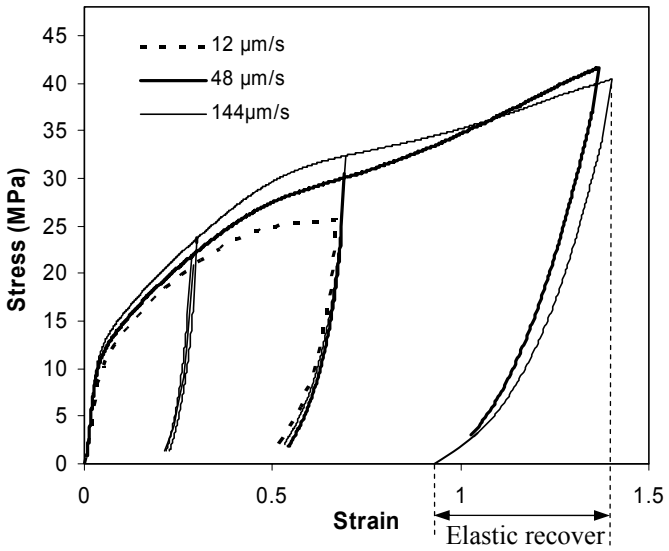

Figure 1: Stress-strain curves of PTFE in compression.

The data in Table 2 show a comparison on the instantaneous elastic recovery under different upsetting conditions. The samples were forged to a half of the original height. For upsetting without dwell time, the lowest speed, i.e. 12 μm/s, gave rise to the smallest amount of elastic recovery. This can be well explained using the single mode Maxwell model, i.e. a spring in series with a dashpot. At slower deformation rates, more energy will be dissipated by the dashpot and less elastic energy will be stored, thus resulting in reduced elastic recovery. With the employment of dwell time, the elastic recovery reduced for all speeds. This again can be explained using the Maxwell model. During the dwell, the stress inside the sample is partially relaxed which in turn produces reduced elastic recovery. The result further indicated a retardation effect at the two higher upsetting speeds with dwell time. It can be seen from the figure that, with the dwell time, the elastic recovery at 48 μm/s is slightly higher than that at 144 μm/s. This cannot be explained using the Maxwell model; however it can be explained using the Kelvin-Voigt model, i.e., a spring in parallel with a dashpot. For the same forging force, the elastic energy story in the Kelvin-Voigt model increases with the decrease of deformation speed. This elastic energy does not give rise to instantaneous elastic recovery if the sample is immediately unloaded following deformation. However, part of this elastic energy will contribute to elastic recovery if a dwell time is applied.

Table 2: Comparison of instantaneous elastic recovery for upset PTFE cylinders with and without dwell under different upsetting speeds.

	12 μm/s	*48 μm/s*	*144 μm/s*
w/ dwell	26.1%	29.1%	27.5%
w/o dwell	31.6%	36.2%	36.9%

The time-dependent dimensional recovery of upset samples without dwell is shown in Figure 2. The strain is in percentage. From the data, it can be seen the post recovery played an important role in determining the final dimension. For example, the post recovery accounted for additionally about 15% strain recovery in the next 6 hours for samples forged at a speed of 12 μm/s. The upsetting speed significantly affected the dimensional recovery process. The lowest speed gave rise to the largest amount of post recovery. This again can be explained using the Kelvin-Voigt model, because slower deformation results in larger strain in the spring. The dwell time also affected the recovery, as shown by the comparison of data between Figure 2 and Figure 3. The amount of post recovery increased with the application of the dwell time. This can also be explained using the Kelvin-Voigt model, as given in the previous paragraph.

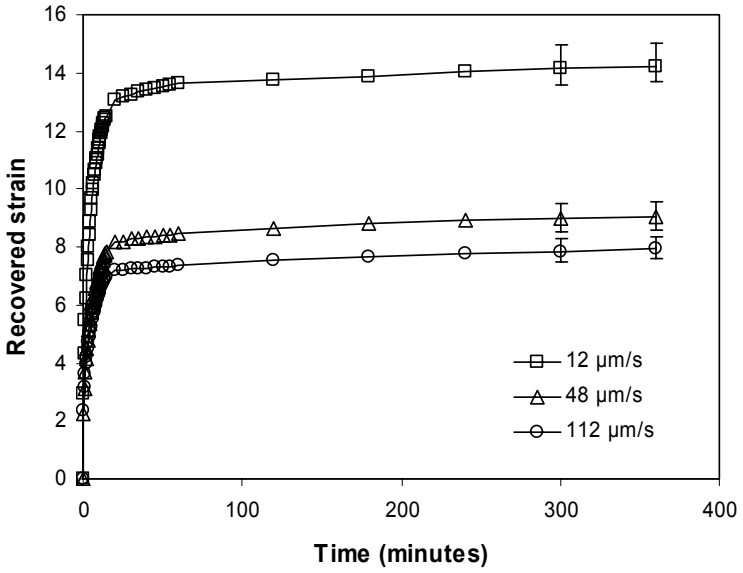

Figure 3: Time-dependent recovery without dwell.

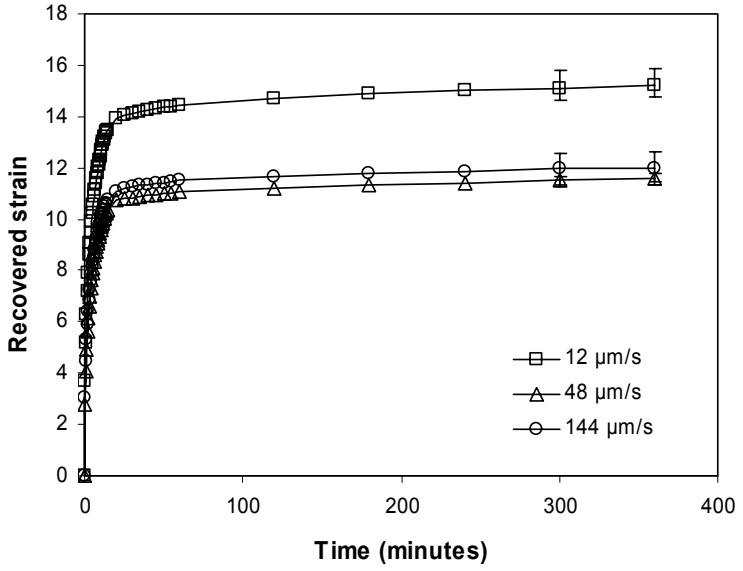

Figure 3: Time-dependent recovery with dwell.

From the above experimental results, it is seen that both the instantaneous elastic recovery and the time-dependent post recovery were significantly affected by the strain applied, the upsetting speed and the dwell time. Either the Maxwell model or the Kelvin model is able to qualitatively explain part of the recovery behavior, but neither alone is able to explain the integrated recovery process. It is thus conjectured that a model with both relaxation and retardation effects may be capable of predicting the entire recovery process observed in the upsetting experiment. One model that considers both the relaxation and retardation effects is a three element Zenar's model [11], as shown in Figure 4. The governing equation for this model is:

$$\frac{E_2}{\eta}\varepsilon + \dot{\varepsilon} = \frac{1}{\eta}\left(1 + \frac{E_2}{E_1}\right)\sigma + \frac{\dot{\sigma}}{E_1} \tag{1}$$

where E is modulus, η is viscosity, σ is stress, and ε is strain. By solving the above equation with boundary conditions, the stress at the end of compression for a specimen compressed under a constant strain rate of $\dot{\varepsilon}$ and with a total strain of ε_0 can be written as:

$$\sigma_{\text{end\_of\_compression}} = \frac{{E_1}^2 \eta \dot{\varepsilon}}{(E_1 + E_2)^2}\left(1 - e^{-\frac{E_1+E_2}{\eta}\frac{\varepsilon_0}{\dot{\varepsilon}}}\right) + \frac{E_1 E_2}{E_1 + E_2}\varepsilon_0 , \tag{2}$$

If a dwell time of t_w is applied immediately after the compression stage, the stress at the end of the dwell can be expressed as (based on the Boltzmann Superposition Principle):

$$\sigma_{\text{end\_of\_dwell}} = \frac{{E_1}^2 \eta \dot{\varepsilon}}{(E_1 + E_2)^2} e^{-\frac{E_1+E_2}{\eta}t_w}\left(1 - e^{-\frac{E_1+E_2}{\eta}\frac{\varepsilon_0}{\dot{\varepsilon}}}\right) + \frac{E_1 E_2}{E_1 + E_2}\varepsilon_0 . \tag{3}$$

After the dwell, the sample is instantaneously unloaded. At the unloading instant, the stress inside the first spring will instantaneously release, resulting in the elastically recovered strain. Therefore, the elastically recovered strain, ε_e, is calculated as:

$$\varepsilon_e = \frac{E_1 \eta \dot{\varepsilon}}{(E_1 + E_2)^2} e^{-\frac{E_1+E_2}{\eta}t_w}\left(1 - e^{-\frac{E_1+E_2}{\eta}\frac{\varepsilon_0}{\dot{\varepsilon}}}\right) + \frac{E_2}{E_1 + E_2}\varepsilon_0 \tag{4}$$

The total strain upon unloading inside the Kelvin part, i.e. ε_2 referring to Figure 4, can also be computed as:

$$\varepsilon_2 = \frac{E_1}{E_1 + E_2}\varepsilon_0 - \frac{E_1 \eta \dot{\varepsilon}}{(E_1 + E_2)^2} e^{-\frac{E_1+E_2}{\eta}t_w}\left(1 - e^{-\frac{E_1+E_2}{\eta}\frac{\varepsilon_0}{\dot{\varepsilon}}}\right). \tag{5}$$

The post strain recovery process can thus be modeled as:

$$\varepsilon_p(t) = \varepsilon_2\left(1 - e^{-\frac{t}{\tau}}\right). \tag{6}$$

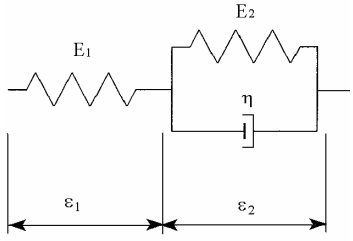

Figure 4: Zernar's model.

The above equations derived from the Zernar's model can be used to qualitatively model the integrated strain recovery process. It can be seen from Equation 3 that the instantaneous elastic recovery with or without dwell increases with the increase of the strain rate. It can also be seen from Equation 3 that an increase of dwell time will result in a decrease of the elastic recovery. Further, from Equation 5 and Equation 6, the post recovered strain with or without dwell decreases with the increase of the strain rate. The actual experiment agreed with the Zernar's model in general except when the upsetting speed increased from 48 μm/s to 144 μm/s with a dwell time. The discrepancy may be attributed to the linearity of the Zernar's model, which is limited to small strain and small strain rate. For large strain and strain rate, nonlinearity needs to be employed.

The Zernar's model described above only provides a qualitative interpretation of the elastic recovery and post recovery processes. The equations derived assumed instantaneous unloading. To incorporate the effect of unloading, a numerical procedure is needed. To accurate model the recovery behavior, nonlinearity needs to be employed in the model, e.g. nonlinear behavior of the springs and the dashpot. The nonlinearity can be modeled phenomenologically or based on molecular

structures. Empirical models and molecular models have been developed in the literature for modeling the flow stress of the material [3]. Similar approaches may be adapted to model the recovery process.

CONCLUSIONS

The deformation and strain recovery characteristics of PTFE during forging and post forging were investigated by a series of upsetting experiments. The dimensional changes after upsetting were found to be a function of upsetting speed, dwell time, and magnitude of strain applied. Samples forged with higher forging speed had higher instantaneous recovery but lower time-dependent recovery. The overall recovery can be reduced by the application of a higher upsetting speed and an appropriate dwell time during forging. The general behavior of PTFE during forging and post forging can be qualitatively predicted using the Zernar's model.

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