

Combining stress relaxation and rheometer test results in modeling a polyurethane stopper[†]

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Abstract

In general, polymers show time-dependent stress-strain responses. The responses can be measured in a time-based test (e.g. a stress relaxation test) or a frequency-based test (e.g. a rheometer test). In a stress relaxation test, it is easy to measure the long-term stress (i.e. the low frequency response) accurately, but it is very difficult, if not impossible, to measure the short-term stress (i.e. the high frequency response) accurately. In contrast, in a rheometer test it is easy to measure the high frequency response by lowering the temperature around the specimen, but it is almost impossible to measure the low frequency response by increasing the temperature over a threshold. Thus, in this paper, a method to combine the two test results was proposed to model a polyurethane stopper for a wide range of frequencies. This method was proven to be valid by showing that simulation results for a drop test and a torque test were in good agreement with test data for a polyurethane stopper used in a robot.

Keywords: Stress relaxation test; Rheometer test; WLF equation; Polyurethane stopper; Hyperelasticity; Viscoelasticity

1. Introduction

Time-dependent material responses of polymers can be measured in a time-based test (e.g. a stress relaxation test) or a frequency-based test (e.g. a rheometer test) [4]. In a stress relaxation test, it is very difficult, if not impossible, to measure the short-term stress (i.e. the high frequency response) accurately because the specimen cannot be deformed fast enough, but it is easy to measure the long-term stress (i.e. the low frequency response) accurately. In contrast, in a rheometer test it is easy to measure the high frequency response by lowering the temperature around the specimen, but it is almost impossible to measure the low frequency response by increasing the temperature because the specimen may be burnt and become a chemically different material. In order to model a polymer stopper used to protect arms in a robot in case of an impact, it is necessary to represent the material response from the low frequency to the high frequency range. To this end, it is a good idea to combine the low frequency response measured in a stress relaxation test and the high frequency response measured in a rheometer test. So far, however, no research has been conducted in modeling a viscoelastic material with consideration of low frequency and high frequency responses obtained from both a stress relaxation test and a rheometer test. Thus,

this paper proposes a method combining the two test results to model a polyurethane (PU) stopper for a wide range of frequencies. The model obtained from this method was applied to a PU stopper used in a robot, and simulation results for a drop test and a torque test were proven to be in good agreement with test data.

2. Materials and experiments

2.1 Hyperelasticity

There are several kinds of analytical models of hyperelasticity: polynomial model ($n = 1\sim 2$), Ogden model ($n = 1\sim 6$), reduced polynomial model ($n = 1\sim 6$), Arruda-Boyce model, Van der Waals model and Marlow model [1]. To formulate the material model of the PU stopper under study, it is essential to determine which types of experiments should be conducted and which model should be used to fit experimental data. To figure out hyperelastic material behavior subjected to various loading modes, the following three types of tests should be conducted [24]: uniaxial compression test (or equibiaxial tension), uniaxial tension test (or equibiaxial compression) and planar (or pure shear) test. If a material is volumetrically deformable, a volumetric test should be also conducted. Since most of rubberlike materials hardly show volumetric deformation, a volumetric test was not conducted in this study. In addition, since the PU stopper is subjected to compression, only the uniaxial compression test was con-

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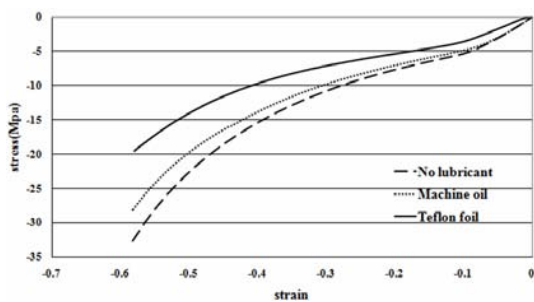


Fig. 1. Uniaxial compression test data using three different lubricants.

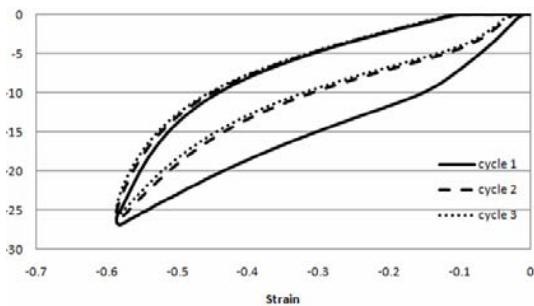


Fig. 2. Mullin's effect.

ducted, and an analytical model of hyperelasticity was selected to fit the compression test data [11].

On the other hand, the following issues should be well considered in dealing with uniaxial compression test data; friction, amount of deformation, Mullin's effect and the maximum strain an analytical model covers [3]. The first issue is that the compression test should be conducted with no friction. Friction entails barreling, which causes the reaction force to rise and also changes a uniaxial compression mode to a multiaxial mode [12].

Fig. 1 shows the uniaxial compression test data obtained by using no lubrication, oil or Teflon foil. There are several methods to reduce friction, but in this study a Teflon foil was used between the specimen and the test jig because it could offer the lowest friction.

The second issue is Mullin's effect, which stems from rearrangement of molecular chains during several initial loading cycles and causes the reduction of the stress, as shown in Fig. 2. However, since the stopper will undergo an impact just once in its lifetime, the first test data was used to determine the model coefficients, i.e., Mullin's effect was not considered in this study. The third issue is a sharp increase of the stress predicted by an analytical model over the maximum strain of the test data as shown in Fig. 3. This means that an analytical model should not be used over the maximum strain measured from tests or tests should be conducted at least up to the maximum strain that occurs in use.

After resolving the issues mentioned above, the uniaxial compression test data was obtained up to strain of -0.75 at a displacement rate of 0.5 mm/sec, and it was fitted by several hyperelastic models using the ABAQUS CAE [1]. Fig. 4(a)-

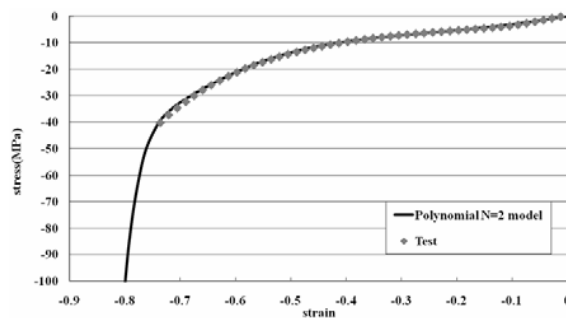


Fig. 3. Stress predicted by a polynomial $N = 2$ model over the maximum strain.

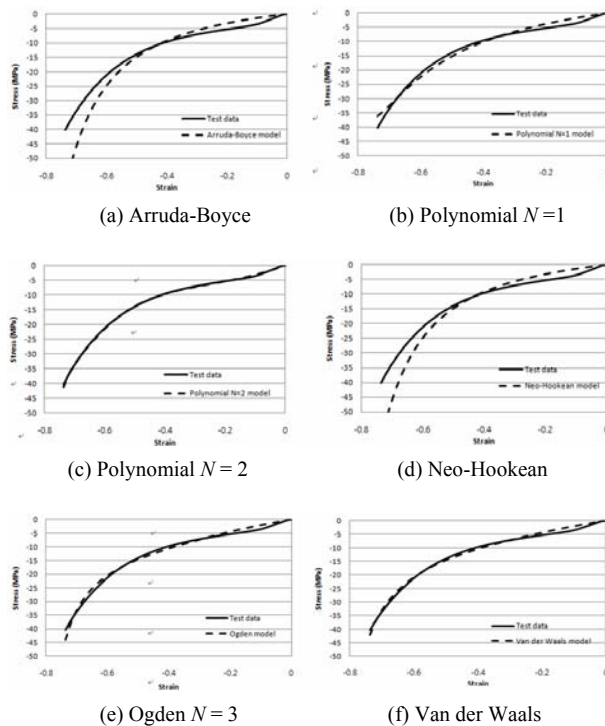


Fig. 4. Measured and predicted stresses.

(f) shows the test data along with fitted curves from several hyperelastic models. The polynomial $n = 2$ model and Ogden $n = 3$ model fit the test data better than the other models, but the polynomial $n = 2$ model is even better than the Ogden $n = 3$ model in high strains, as shown in Fig. 5. In addition, the coefficients of the Ogden model are not unique. In other words, there exist several sets of coefficients that fit a nominal stress-strain curve, and they do not have similar values for similar nominal stress-strain curves [5]. Thus, it was decided to use the polynomial $n = 2$ model as a hyperelastic model in this study. Which hyperelastic model fits well depends on each polymer [20]. Polynomial $n = 2$ model is the best choice for the PU stopper under study, but the reduced polynomial $n = 3$ model (=Yeoh), Van der Waals model or Arruda-Boyce model could be the best choice for other polymers [13].

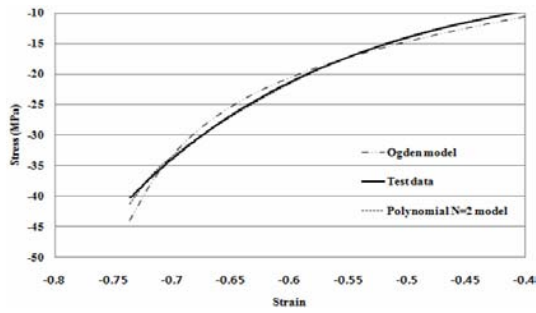


Fig. 5. Test vs. Polynomial N = 2 and Ogden model.

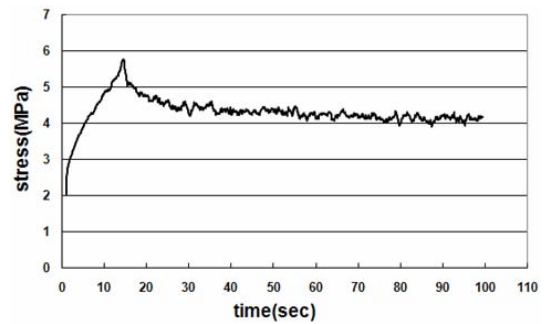


Fig. 6. Stress relaxation test data.

2.2 Viscoelasticity measured by stress relaxation and rheometer tests

The viscoelastic model represented by the Prony series has been widely used to model time-dependent behavior of polymers [23]. The analytical form shown in Eq. (1) in the time domain can be Laplace-transformed to the form in the frequency domain shown in Eq. (2) [17].

$$G(t) = G_0 \left\{ 1 - \sum_{i=1}^M g_i \left(1 - e^{-t/\tau_i} \right) \right\} \tag{1}$$

$$G'(\omega) = G_0 \left[1 - \sum_{i=1}^M g_i \right] + G_0 \sum_{i=1}^M \frac{g_i \tau_i^2 \omega^2}{1 + \tau_i^2 \omega^2} \tag{2}$$

$$G''(\omega) = G_0 \sum_{i=1}^M \frac{g_i \tau_i \omega}{1 + \tau_i^2 \omega^2}$$

Here, G_0 is the short-term shear modulus, (g_i, τ_i) are the Prony series parameters, $G_0 \left(1 - \sum_{i=1}^M g_i \right)$ represents the long-term shear modulus G_∞ , $G'(\omega)$ is the storage modulus, $G''(\omega)$ is the loss modulus, and M is the number of the Prony series parameters. There are two kinds of experiments to measure the time-dependent behavior; one is a stress relaxation test (or creep test, but in this study a stress relaxation test was conducted) which results in a shear modulus as a function of time [6], and the other is a rheometer test which results in the storage modulus and the loss modulus as a function of frequency. Each test has advantages and disadvantages, but now a rheometer test seems to be more popular because it is more reliable and easier to conduct [22]. In a stress relaxation test it is very difficult, if not impossible, to measure G_0 accurately because the maximum strain rate of a tester is not high enough, but it is easy to measure G_∞ accurately. In a rheometer test it is easy to measure the storage and loss moduli for a comparatively narrow range of frequencies (e.g. from 5 Hz to 30 Hz) under various temperatures. By using Williams-Landel-Ferry (WLF) equations, the storage and loss moduli can be shifted to those at a reference temperature, which is usually the ambient temperature [21]. The moduli measured at lower temperatures will be shifted to those at higher frequencies at the reference temperature, and the moduli measured at higher temperatures will be shifted to those at lower frequencies at the reference

temperature. However, it is not appropriate to raise the temperature over a threshold because the material may be burnt and its properties may become quite different [14]. Thus, it is difficult to obtain the material response in frequency range lower than 0.001 Hz at the reference temperature although it is easy to measure the material response in high frequency range. It is noteworthy that the response in the beginning stage of a stress relaxation test is related to that in the high frequency range of a rheometer test, and the response in the ending stage of a stress relaxation test is related to that in the low frequency range of a rheometer test. In other words, the two test data can complement each other.

A stress relaxation test was conducted at a strain rate of 4/sec based on ASTM D-1646, and its result is shown in Fig. 6. Note that the stress did not start from a certain value at time 0 and it was impossible to determine the short-term shear modulus G_0 from the stress relaxation test. A rheometer test was conducted at every 10°C from -10°C to 80°C using DMA (Dynamic Mechanical Analyzer) based on ASTM D-5026, and its result is shown in Fig. 7. Using the WLF equation shown in Eq. (3), the master curves for the storage modulus and the loss modulus were obtained as shown in Fig. 8 [10].

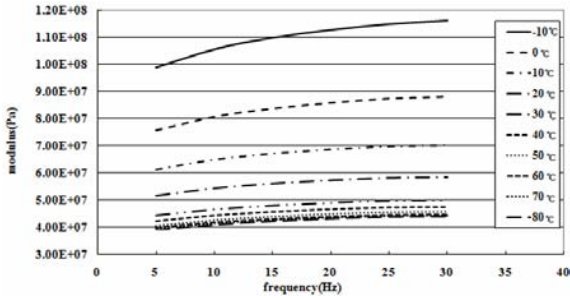
$$C_1 = \frac{C_1^g}{1 + (T_r - T_g) / C_2^g} \tag{3}$$

$$C_2 = C_2^g + T_r - T_g$$

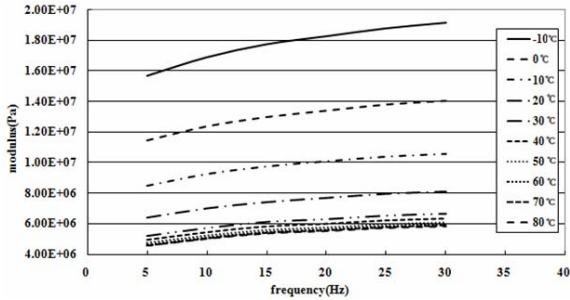
$$a_T = 10^{\left(\frac{-C_1(T - T_r)}{C_2 + T - T_r} \right)}$$

Here, C_1^g and C_2^g are universal constants (the values are 17.44 and 51.6, respectively, but Van Krevelen recommended 8.86 and 101.6, respectively [19].), T_r and T_g are the reference temperature of 293K and the glass transition temperature of 238K of PU, respectively [Hepburn, 1982].

In order to determine the Prony series parameters that fit the storage and the loss modulus over a wide range of frequencies, an error measure to minimize should be defined [2, 15]. A mean square error is usually used as an error measure shown in Eq. (4) [1], but in this study another error measure was defined as shown in Eq. (5). Rubber components such as a PU stopper are usually used as a damper of which the damping characteristic can be related to the ratio of the loss modulus to



(a) Storage modulus



(b) Loss modulus

Fig. 7. Storage modulus and loss modulus measured at different temperatures.

the storage modulus, so-called $\tan \delta$. Thus, the error measure to minimize the difference between the measured $\tan \delta$ and the analytical form of $\tan \delta$ from Eq. (2) was used in this study.

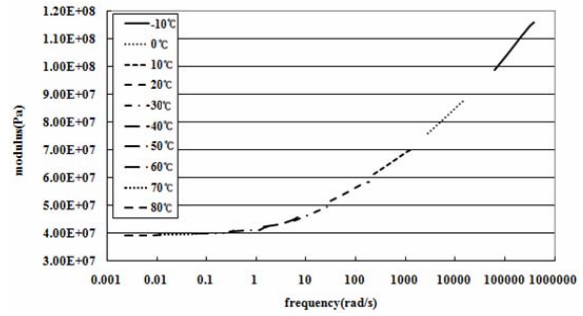
$$Error = \sum_{i=1}^N \frac{1}{G_{\infty}^2} \left[(G' - G'_{test})_i^2 + (G'' - G''_{test})_i^2 \right] \quad (4)$$

$$Error = \sum_{i=1}^N \left[\left(\frac{G''}{G'} \right)_i - \left(\frac{G''_{test}}{G'_{test}} \right)_i \right]^2 \quad (5)$$

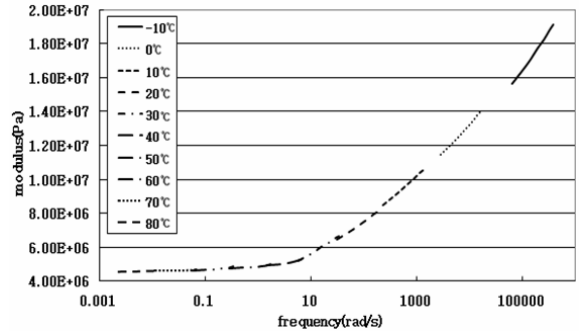
Here, G' or G'' is the storage or the loss modulus analytically defined in Eq. (2) and G'_{test} or G''_{test} is the storage or the loss modulus obtained from the rheometer test and the WLF equation, which is shown in Fig. 8. In addition, N is the number of values taken from Eq. (2) and Fig. 8.

The ratio of the loss modulus to the storage modulus is shown for a wide range of frequencies in Fig. 9. In order to obtain the ratio at very low frequency the rheometer test should be conducted at high temperature. However, it is not appropriate to increase the temperature over a threshold (e.g. 80°C for PU) because the specimen may be burnt and its mechanical properties may become unrepresentative. In order to obtain the ratio for very low frequency the curve may be extrapolated with caution.

Once the curve for the ratio is obtained, there is still an issue left: which frequency range should be fitted to accurately capture the viscoelastic behavior. If the low frequency test



(a) Storage modulus



(b) Loss modulus

Fig. 8. Storage modulus and loss modulus as a function of frequency.

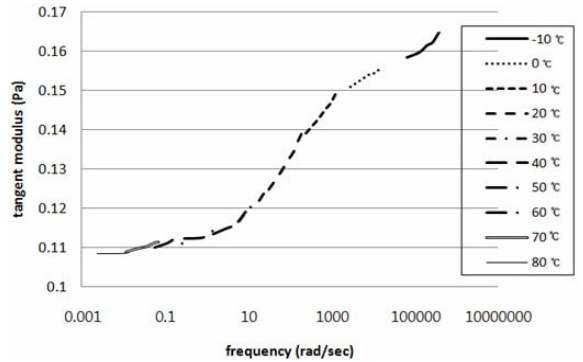


Fig. 9. $\tan \delta$ as function of frequency.

data are fitted closely, a slow (or static) deformation can be described well. However, if the high frequency test data are fitted closely, a fast (dynamic) deformation can be described well. The Prony series parameters were determined to minimize the error measure defined in Eq. (5), using the ratio for four different lower bound frequencies of 0.001 Hz, 0.01 Hz, 0.1 Hz and 1 Hz with a fixed upper bound frequency of 1000 Hz. The shear modulus obtained from Eq. (1) using the Prony series parameters is plotted in time domain in Fig. 10. Note that the shear modulus determined based on the test data from 0.01 Hz to 1000 Hz matches well with the stress relaxation test data in the long term, but the shear modulus determined based on the test data for other frequency ranges shows some difference from the stress relaxation test data in the long term.

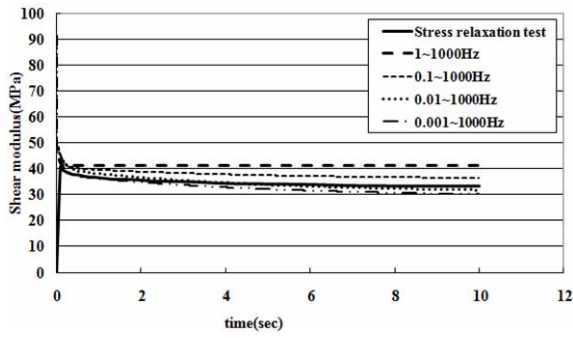


Fig. 10. Shear modulus determined based on four different lower bound frequencies vs. stress relaxation test data.

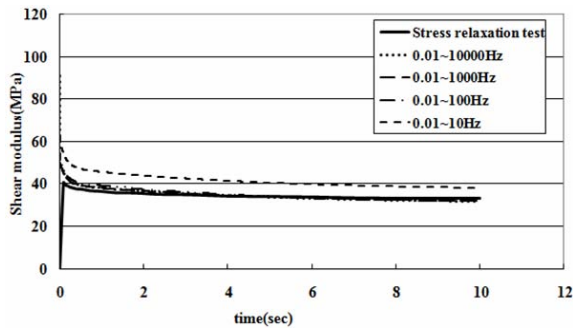


Fig. 11. Shear modulus determined based on four different upper bound frequencies vs. stress relaxation test data.

In addition, the Prony series parameters were determined to minimize the error measure defined in Eq. (5), using the ratio for four different upper bound frequencies of 10 Hz, 100 Hz, 1000 Hz and 10000 Hz with a fixed lower bound frequency of 0.01 Hz. The shear modulus obtained from Eq. (1) using the Prony series parameters is plotted in time domain in Fig. 11. Note that the upper bound frequency over 100 Hz does not change the short-term and the long-term shear modulus significantly, and it was determined to use 1000 Hz as the upper bound for the frequency range.

3. Validation of material model

A compression specimen of which the diameter is 30 mm and the height is 19 mm was made, and a ram was dropped to the compression specimen at three different heights, resulting in an impact speed of 3.1, 4.3 or 5.9 m/s. An FE model shown in Fig. 12 was created using the polynomial $n = 2$ hyperelastic model and viscoelastic model with the Prony series parameters determined in the previous section. The test data and simulation results on the load vs. time were shown in Fig. 13. Note that the simulation results were in good agreement with the test data for three different impact speeds.

A robot with the stopper installed was also modeled, and its FE model was created as shown in Fig. 14. An arm of the robot was torque controlled, and the torque vs. the rotated angle was measured until the stopper was severely deformed.

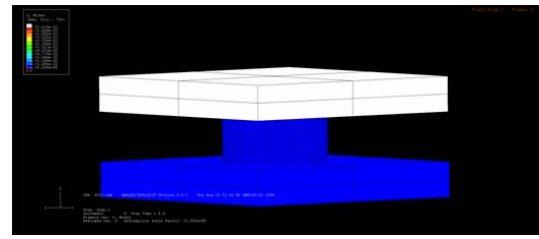


Fig. 12. FE model for the drop test.

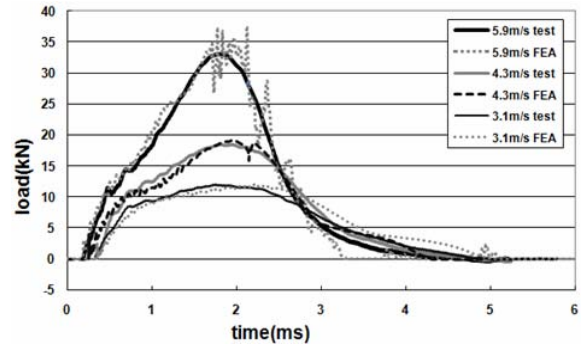


Fig. 13. Simulation and drop test results at three different speeds.

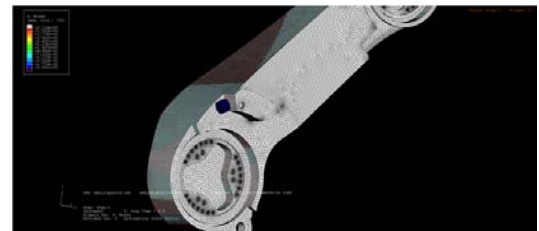


Fig. 14. Robot arm and a PU stopper FE model.

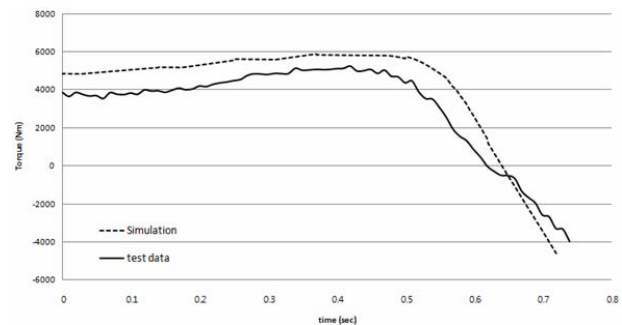


Fig. 15. Simulation and test results on torque-angle relation.

A simulation using the FE model was also conducted, and its torque vs. the rotated angle was compared with the test data as shown in Fig. 15, showing that the simulation result was in good agreement with the test data. In addition, the deformed shaped of the stopper obtained from the simulation was overlaid on the photo of the deformed stopper in the robot, and it was noticed that the deformed shape from the simulation was also in good agreement with that captured in the photo.

4. Conclusions

From the simulation and test results mentioned in this study, the following conclusions can be reached.

(1) For the PU stopper under study, the polynomial $n = 2$ model was the best model to fit the compression test data.

(2) An error measure for $\tan \delta$ was used to determine the Prony series parameters.

(3) A frequency range of 0.01 Hz to 1000 Hz resulted in the best correlation with the long-term shear modulus obtained from a shear relaxation test.

(4) The hyperelastic model and the viscoelastic model determined in this study resulted in good agreement with the compression tests for three different drop speeds and with the torque-angle test for the PU stopper used in a robot.

(5) Thus, the methodology of determining hyperelastic and viscoelastic models in such a way that the short term modulus is determined from the rheometer test and the long term modulus is determined from the stress relaxation test results in a reasonable material model for a rubberlike stopper.

Acknowledgment

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References

- [1] ABAQUS, 2007, ABAQUS Theory Manual, version 6.5-1.
- [2] J. Bergström, Ph.D., 2005, *Calculation of Prony Series Parameters From Dynamic Frequency Data*.
- [3] M. C. Boyce, S. Socrate and P. G. Llana, Constitutive model for the finite deformation Stress-Strain behavior of poly(ethylene terephthalate) above the glass transition, *Elsevier*, 41 (6) (1999) 2183-2201.
- [4] T. Chen, *Determining a Prony Series for a Viscoelastic Material from Time Varying Strain Data*, US Army Research Laboratory Vehicle Technology Directorate Langley Research Center Hampton Virginia, 2000.
- [5] J. H. Choi, H. J. Kang, H. Y. Jeong, T. S. Lee and S. J. Yoon, Heat Aging Effect on the Material Property and the Fatigue Life of Vulcanized Natural Rubber and Fatigue Life Prediction Equation, *Journal of Mechanical Science and Technology*, 19 (2005) 1229-1239.
- [6] T. Dalrymple, J. Choi and K. Miller, Elastomer Rate-Dependence: A Testing and Material Modeling Methodology, Axel Products Inc, 172nd Technical Meeting of the Rubber division of the American Chemical Society (2007) 1547-1977.
- [7] C. Hepburn, 1982, *Polyurethane elastomers*, *Applied Science*, London.
- [8] M. Hjiij, G. de Saxce and Z. Mroz, Influence of frictional anisotropy on contacting surfaces during Loading/Unloading cycles, *International Journal of Non-Linear Mechanics*, 41 (2006) 936-948.
- [9] V. H. Kenner, B. D. Harper and V. Y. Itkin, Stress relaxation in molding compounds, *Journal of Electronic Materials*, 26 (1997) 821-826.
- [10] A. S. Khan and O. Lopez-Pamies, Time and Temperature dependent response and relaxation of a soft polymer, *International Journal of Plasticity*, 18 (2001) 1359-1372.
- [11] B. Magnain and J. M. Cros, Solution of large deformation impact problems with friction between blatz-ko hyperelastic bodies, *International Journal of Engineering Science*, 44 (2006) 113-126.
- [12] J. Michael, Unilateral compression of rubber, *Journal of Applied Physics*, 26 (1955) 1104-1106.
- [13] R. W. Ogden and D. G. Roxburgh, A Pseudo-elastic model for the mullins effect in filled rubber, The Royal Society, Mathematical, *Physical and Engineering Science*, 455 (1998) 2861-2877.
- [14] L. Y. Robert and L. Suckhong, Short-term and Long-term aging behavior of rubber modified asphalt paving mixture, *Transportation Research Record*, 1530 (1996) 11-17.
- [15] R. J. Scavuzzo, Oscillating stress on viscoelastic behavior of thermoplastic polymers, *Journal of Pressure Vessel Technol.*, 122 (2008) 386-389.
- [16] D. W. Schaffner, The application of the wlf equation to predict lag time as a function of temperature for three psychrotrophic bacteria, *International Journal of Food Microbiol.*, 27 (1994)107-115.
- [17] M. L. Slanik, J. A. Nemes, M. J. Potvin and J. C. Piedboeuf, Time domain finite element simulations of damped multilayered beams using a prony series representation, *Mechanics of Time-Dependent Materials*, 4 (2000) 211-230.
- [18] K. W. Song and G. S. Chang, Rheological Behavior of Viscoelastic Semi-Solid Ointment Base (vaseline) in Oscillatory Shear Flow Fields, *Journal of Pharmaceutical Investigation*, 36 (2006) 31-38.
- [19] L. C. E. Strulk, DSM Research, On the Van Krevelen / Hoftyzer Relationship for the High-Temperature Limiting Viscosities of Polymer Melts, *Polymer*, 38 (1997) 1477-1479.
- [20] C. Vallee, D. Fortune and F. Peyraut, The 3^e hyperelastic model applied to the modeling of 3D impact problems, *Journal of Finite Element in Analysis and Design*, 43 (2006) 1927-1941.
- [21] M. L. William, R. F. Landel and J. D. Ferry, The Temperature dependence of relaxation mechanisms in amorphous polymer and other glass-forming liquids, *Journal of American Chem. Soc.*, 77 (1955) 3701-3706.
- [22] H. H. Winter, Analysis of Dynamic mechanical data: inversion into a relaxation time spectrum and consistency check, *Journal of Non-Newtonian Fluid Mechanics*, 68 (1996) 225-239.
- [23] L. M. Yang, V. P. W. Shim and C. T. Lim, A visco-hyperelastic approach to modelling the constitutive behaviour of rubber, *International Journal of Impact Engineering*, 24 (1999) 545-560.
- [24] J. Zhang, N. Kikuchi, V. Li, A. Yee, and G. Nusholtz, Constitutive modeling of polymeric foam material subjected

to dynamic crash loading, *International Journal of Impact Engineering*, 21 (1997) 369-386.



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