Material Modeling of Polylactide

Senthil K. Eswaran[1], J. Ashley Kelley[1], Jorgen S. Bergstrom[2], Virginia L. Giddings[1]

[1] Abbott Vascular, Santa Clara, CA 95054

[2] Veryst Engineering, Needham, MA 02494

Abstract: Computational modeling of stents can provide insight into critical locations (high stress/strain regions), help with design iterations/optimization, and reduce the need for bench-top testing. This study focuses on the developmental efforts to create a material model that can capture the mechanical response of poly-L-lactide (PLLA), the backbone of Abbott Vascular's ABSORB Bioresorbable Vascular Scaffold (BVS). PLLA is an anisotropic, viscoplastic material. Its material behavior maybe affected by temperature, environment, and processing; thereby, introducing substantial challenges in modeling its response. A new test protocol was developed for performing material characterization experiments that can capture the anisotropic material response using a dog-bone shaped specimen. The Parallel Network (PN) model from Veryst Engineering is an advanced modular constitutive model framework. The PN model allows for specific anisotropic and isotropic hyperelastic and viscoplastic components to be combined to capture the experimentally observed response of PLLA. The material model (PN model) was calibrated to experimental data from the dog-bone material testing. A validation experiment (single ring tensile test) was performed and the fidelity of the computational model was tested by comparing its predictions to the single ring tensile test data.

Keywords: Stent, Anisotropy, Viscoelasticity, Viscoplasticity, Polylactide, Constitutive model, Parallel network model, Dog-bone material testing, Calibration, Validation.

1. Introduction

The role of computational modeling in medical devices is becoming increasingly important since it can provide insight into critical locations of the device (high stress/strain regions), help with design iterations/optimization, and reduce the need for bench-top testing; thereby, reducing the cycle time during the development of the device. Modeling of medical devices poses substantial challenges in terms of accurately capturing the material behavior under relevant processing and/or physiological conditions, and modeling of the *in vivo* loading/boundary conditions. Developing a validated computational model in this context is critical to achieving robust model predictions.

This paper focuses on the developmental efforts to create a material model that can capture the mechanical response of poly-L-lactide (PLLA), the backbone of Abbott Vascular's (Santa Clara, CA) ABSORB bioresorbable vascular scaffold (BVS). Polylactide polymers have a long history of use in the medical device industry starting with bioresorbable sutures in 1960s (Amass, 1998). PLLA is a biodegradable, semi-crystalline thermoplastic that is being increasing used in medical devices due to its biodegradability, processability, and biocompatibility (Amass, 1998; Ikada, 1999). PLLA is an anisotropic (processing-dependant), viscoplastic material and its material behavior maybe affected by temperature, environment, and processing (Grabow, 2005; Engelberg,

Visit the SIMULIA Resource Center for more customer examples.

1991; Perego, 1996; Garlotta, 2001). Given the dependence of the material response on such factors, it is particularly relevant that characterization of the material response in the context of the specific application is performed.

The material response of PLLA is characterized by rate-dependant anisotropic viscoplasticity. These characteristics inhibit the use of simple material models, such as metal plasticity and linear viscoelasticity. Instead, in order to perform accurate FE model predictions it is necessary to use an advanced user-material model in order to capture the true response of PLLA. In this work, the Parallel Network (PN) model has been used. The PN model is a modular constitutive model framework that has been developed by Veryst Engineering (Needham, MA) and is commercially available for Abaqus Standard and Explicit.

The objective of this study was to develop a validated material model for PLLA that can be used for modeling of scaffolds. The experimental characterization of the material behavior, constitutive model theory, calibration and validation of the material model are presented in this paper.

2. Material Characterization

Tensile testing using dog-bone shaped specimen is a widely used test methodology that provides stress-strain response of the material under investigation. It is critical that the specimen used in the tensile tests consists of the material that is representative of the finished good product. Thus, if there are any manufacturing processes that can affect the material response of the finished good product, the dog-bone specimens should be subjected to those manufacturing processes or equivalent conditions.

To accurately capture the anisotropy and the relevant manufacturing processes on a micro-specimen scale, new fixtures were developed to test dog-bone shaped specimens that are laser-cut from the PLLA tubing in both the hoop and axial directions. The dog-bone specimens had the recommended geometry for polymer testing of samples less than 4 mm in thickness as specified in ASTM D638. The samples were scaled by matching the ASTM “width of the narrow section” to approximately one strut width in the scaffold design.

The uniaxial experimental study involved monotonic loading to failure in the hoop and the axial directions at three different applied displacement rates (0.01 in/min, 0.10 in/min, 1.00 in/min). The experimental tests for each condition were repeated 10 to 15 times for each test speed to obtain the statistical distribution of the stress-strain behavior. The raw experimental data was corrected to compensate for the compliance of the load train (Xiao, 2008). For each condition, the experimental data curve that is closest to the average of all curves at that condition was selected as the representative curve. Only the representative curves were used for the stress-strain material model calibration.

Figure 1 shows the stress-strain curves that were used for the material model calibration. These curves show that the material is highly anisotropic after yield: the flow resistance in the hoop (circumferential) direction continues to grow with strain after yield, but the response in the axial (longitudinal) direction is significantly softer and exhibits necking. The material response is also strain-rate dependent, as is typically seen for thermoplastics.

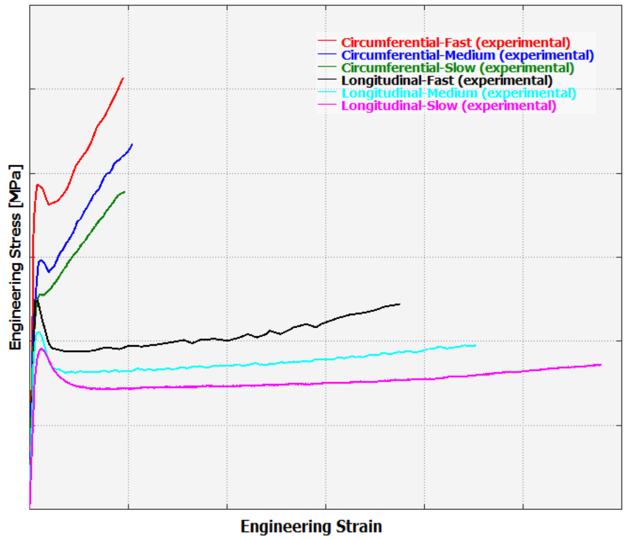


Figure 1. Uniaxial experimental data.

3. Constitutive Model Theory

The anisotropic viscoplastic response of the PLLA can be captured by representing the microstructure as two molecular networks that act in parallel, see Figure 2. The first network (*A*) consists of an isotropic neo-Hookean model with viscoplastic flow driven by stress-based activation energy. The second network (*B*) consists of an anisotropic hyperelastic model that captures the large strain behavior in different directions.

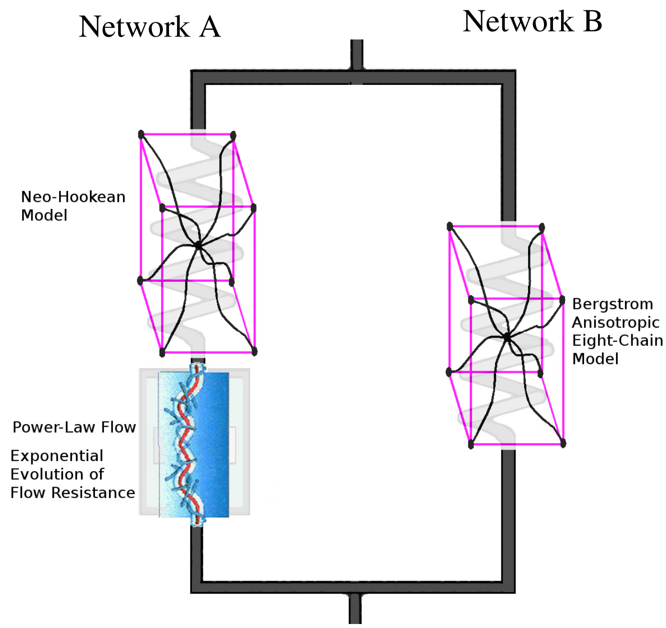


Figure 2. Rheological representation of the Parallel Network model used to capture the response of the PLLA.

The constitutive equations for this model are similar in structure and derivation to existing models [Bergstrom, 2010; Chakrabarti-Bell, 2010; Bergstrom, 2001], but the specific details presented below are unique to PLLA.

In the material model the total deformation gradient is multiplicatively decomposed into elastic and viscoplastic components: $\mathbf{F} = \mathbf{F}_A^e \mathbf{F}_A^v = \mathbf{F}_B$.

The Cauchy stress in Network *A* is given by a Neo-Hookean model:

$$\boldsymbol{\sigma}_A = \frac{\mu_A}{J_A^e} \mathrm{dev}[\mathbf{b}_A^{e*}] + \kappa (J_A^e - 1)\mathbf{I} \tag{1}$$

where μ_A is the shear modulus, $J_A^e = \det(\mathbf{F}_A^e)$, $\mathbf{F}_A^e$ is the elastic part of the deformation gradient($\mathbf{F}$), $\mathbf{b}_A^{e*} = (J_A^e)^{-2/3} \mathbf{F}_A^e (\mathbf{F}_A^e)^T$ is the distortional left Cauchy-Green tensor, and κ is the bulk modulus.

The rate of viscoplastic flow of the material is given by the rate of deformation tensor:

$$\tilde{\mathbf{D}}_A^v = \left(\frac{\tau}{f_\varepsilon \tau_b}\right)^m \frac{\mathrm{dev}[\boldsymbol{\sigma}_A]}{\tau} \tag{2}$$

where $\tau = (\mathrm{tr}[\sigma'_\mathrm{A}\sigma'_\mathrm{A}])^{1/2}$ is the driving shear stress, σ'_A is the deviatoric part of σ_A ,

$$f_\varepsilon = f_f + (1 - f_f)\exp\left[\frac{-\varepsilon_A^v}{\varepsilon_h}\right] \tag{3}$$

$$\varepsilon_A^v = ||\ln \mathbf{v}_B^v|| \tag{4}$$

and where $\tau_b, m, f_f, \varepsilon_h$ are material parameters and $\mathbf{v}_\mathrm{B}^\mathrm{v}$ is the viscoelastic stretch tensor of Network *A*.

The Cauchy stress in Network *B* is given by the Bergstrom anisotropic eight-chain model [Chakrabarti-Bell, 2010]. The experimental data indicates that the PLLA material is stronger in the circumferential direction than in the axial direction. The response in the third material orientation (radial direction) is assumed to be identical to the axial response. Under these conditions the stress is given by:

$$\boldsymbol{\sigma}_B = \frac{\mu_B}{J\lambda_c}\frac{L^{-1}(\lambda_c/\lambda_L)}{L^{-1}(1/\lambda_L)}\mathrm{dev}[\mathbf{b}^*] + \kappa(J-1)I + \frac{B}{\lambda_f}\left(1 - \frac{1}{\lambda_f}\right)\mathbf{Fe}_2 \otimes \mathbf{Fe}_2 \tag{5}$$

where $L^{-1}(x)$ is the inverse Langevin function (Arruda, 1993), $\lambda_c = (\mathrm{tr}[\mathbf{b}^*]/3)^{1/2}$ is the chain stretch, $\mathbf{b}^* = J^{-2/3}\mathbf{F}\mathbf{F}^\mathrm{T}$, $J = \det[\mathbf{F}]$, $\lambda_f = \|\mathbf{Fe}_2\|$ is the molecular fiber stretch, κ is the bulk modulus, and μ_B, *B* are material parameters.

The total Cauchy stress is given by the sum of the stresses in Network *A* and the stress in Network *B*: $\sigma = \sigma_A + \sigma_B$.

4. Material Model Calibration

The PN model was calibrated to the uniaxial experimental data summarized in Section 2. The calibration was performed using the procedure depicted in Figure 3. First an initial estimate of the material parameters was determined based on experience with other similar materials. Then the material model with the specified set of parameters was used to predict the response of the uniaxial experiments. The coefficient of determination, R^2, was then used to evaluate how close the model predictions were to the experimental data. The Nelder-Mead Simplex algorithm (Nelder, 1965) then automatically adjusted the material parameters and repeated the cycle until an optimal fit between the experimental data and the model predictions is obtained.

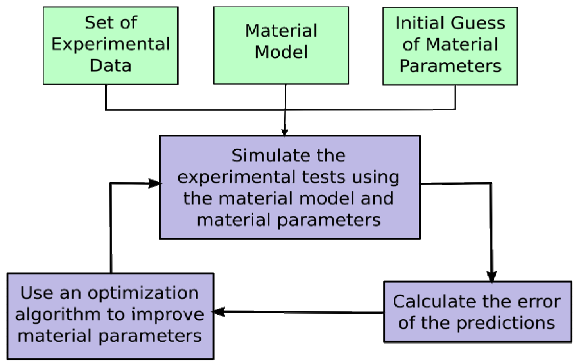


Figure 3. Schematic illustrating the calibration procedure used to determine the material parameters in the Parallel Network model.

The calibration procedure was performed using the MCalibration software that is commercially available from Veryst Engineering. Comparisons between the experimental stress-strain data and the calibration predictions are shown in Figure 4. The results show that the anisotropic, rate-dependant response of the material has been captured using this material model.

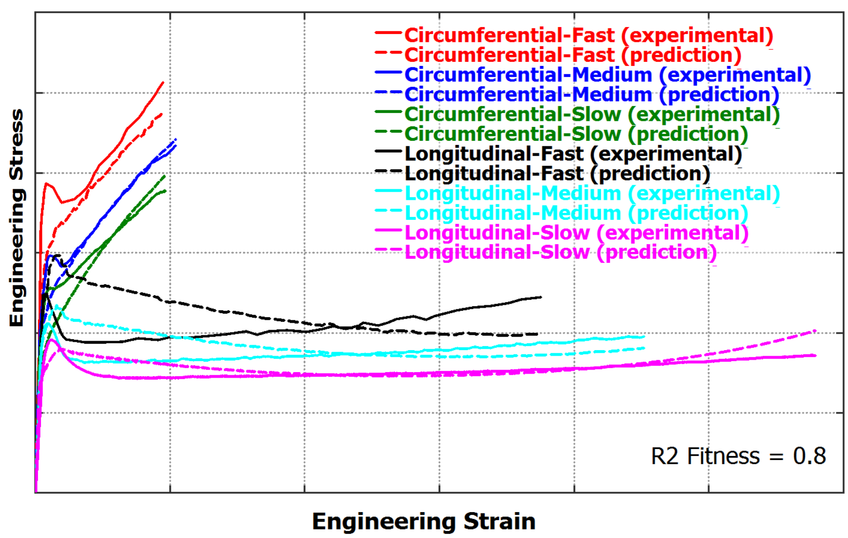


Figure 4. Comparison between the uniaxial experimental data and the calibration predictions.

5. Model Validation

5.1 Ring Tension Experiment

The PLLA material was tested using a ring tension experiment (Figure 5) to provide experimental data for model validation. In these experiments, a single ring of the scaffold was pulled in tension between two cylindrical rods. The applied displacement rate was 0.1 inch/minute (0.042 mm/sec). Both monotonic loading to failure and cyclic load-unload type experiments were performed. Each experimental ring tension condition was repeated using 3-4 scaffolds and testing 5-9 rings per scaffold to obtain the distribution in force-displacement behavior. Figure 6 shows the average force-displacement data from the ring tension experiments.

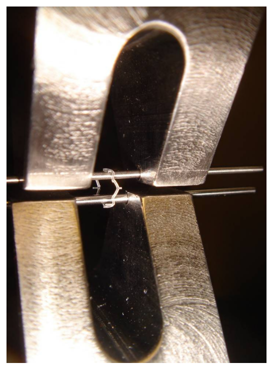


Figure 5. Photograph showing the experimental setup used in the ring tension experiment.

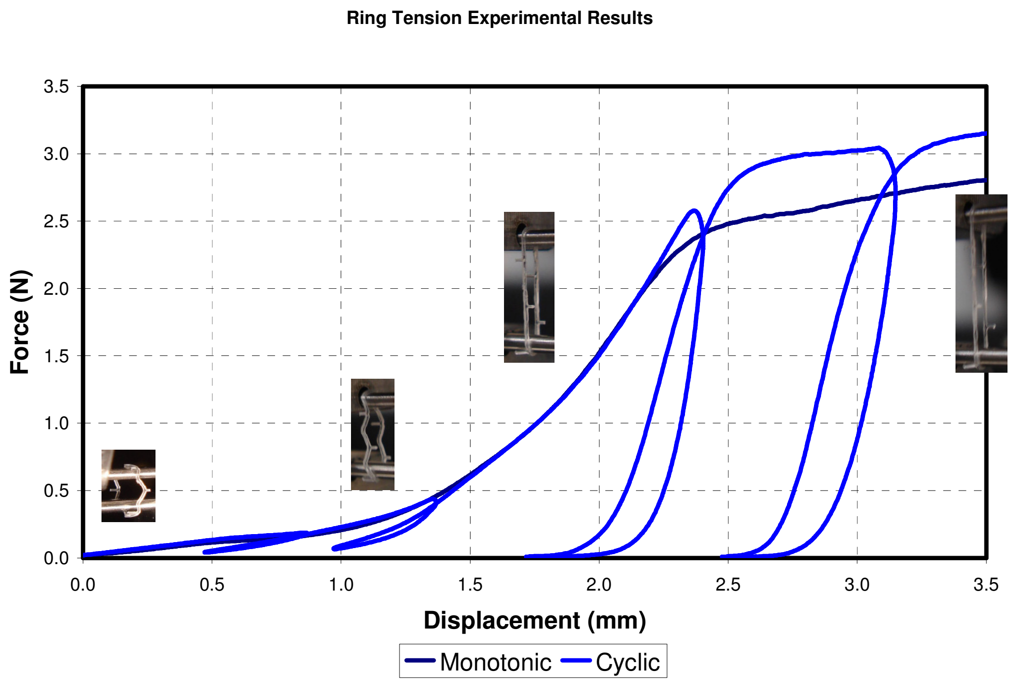


Figure 6. Average force-displacement curves from the ring tension experiments.

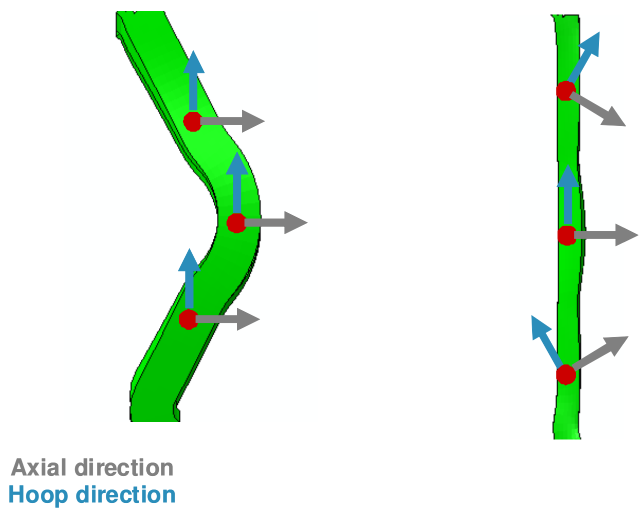
5.2 FE Model Predictions

A FEA model of a single ring of the scaffold was developed to simulate the ring tension experiment. Figure 7 shows the FE mesh used to represent the geometry in the ring tension test. The bottom pin was held fixed and the top pin was displaced upwards using the same displacement rate as was used in the experimental tests. The top and bottom pins were modeled as analytical rigid cylinders. The FE simulations were performed using Abaqus/Standard 6.10. Two types of FE simulations were performed: monotonic loading to failure, and cyclic loading to failure with the unloading starting at displacement magnitudes that match the experimental data. Figure 8 shows the undeformed and deformed shape of a single crest with the corresponding material orientation. It is clear that both axial and hoop properties contribute to the tensile force during the single ring test, thereby providing a validation for the anisotropic material properties.

The results from the FE simulations are compared to the experimental data in Figure 9. The figure shows that the predicted force-displacement behavior is in excellent agreement with the experimental data for both the monotonic and load-unload tests.

Undeformed configuration *Deformed configuration*

Figure 7. FE mesh used to simulate the ring tension experiment, showing initial and final configurations.



Undeformed shape *Deformed shape*

Figure 8. Undeformed and deformed shape of a single crest indicating that both axial and hoop properties contribute to the tensile force during the ring tension test.

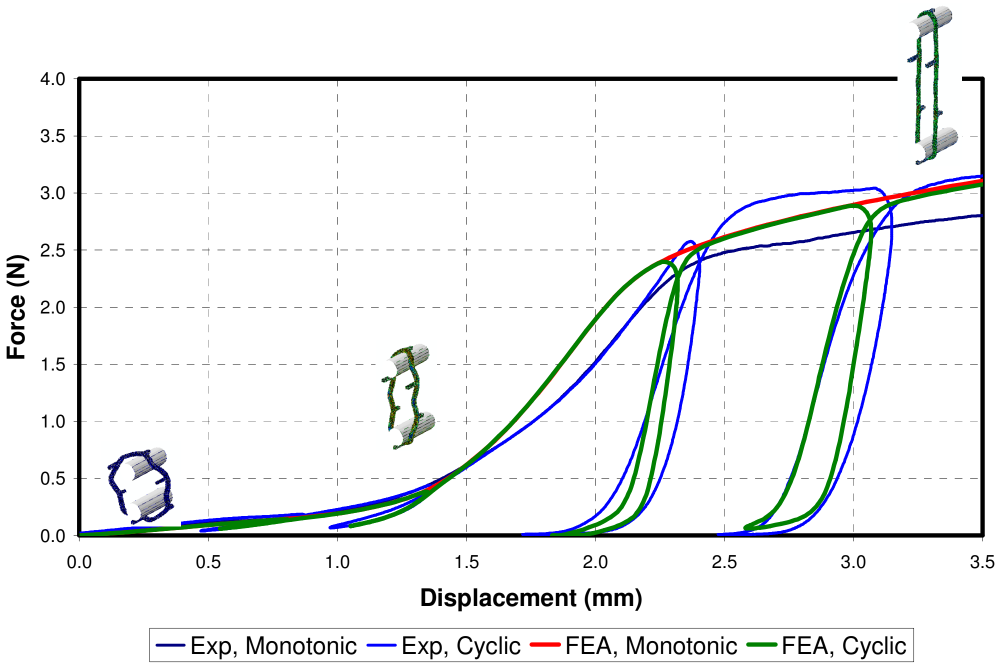


Figure 9. Comparison between the experimental ring-tension data and predictions from FE simulations using the material model for PLLA.

6. Summary

A material model that captures the anisotropic, viscoplastic material behavior of PLLA was developed and validated. The material behavior of PLLA was characterized using a new test protocol that allowed the characterization of the anisotropic material properties with the specimens experiencing relevant manufacturing process conditions. The parallel network material model was calibrated to the experimental uniaxial data. The model predictions were then validated using the ring tension experimental data with the results showing very favorable comparison between model and experiment.

7. References

1. Amass, W., Amass, A., and Tighe, B., “A Review of Biodegradable Polymers: Uses, Current Developments in the Synthesis and Characterization of Biodegradable Polyesters, Blends of Biodegradable Polymers and Recent Advances in Biodegradation Studies,” Polymer International, vol. 47, pp. 89-144, 1998.

2. Arruda, E.M., and Boyce, M.C., “A Three-Dimensional Constitutive Model for the Large Stretch behavior of Rubber Elastic Materials”, Journal of the Mechanical and Physics of Solids, vol. 41, pp. 389-412, 1993.
3. ASTM D639, “Standard Test Method for Tensile Properties of Plastics,” 2003.
4. Bergstrom, J.B., and Bischoff, J.E., “An Advanced Thermomechanical Constitutive Model for UHMWPE,” International Journal of Structural Changes in Solids, vol. 2, pp. 31-39, 2010.
5. Bergstrom, J.B., and Boyce, M.C., “Constitutive Modeling of the Time-dependent and Cyclic Loading of Elastomers and Application to Soft Biological Tissues,” Mechanics of Materials, vol. 33, pp. 523–530, 2001.
6. Chakrabarti-Bell, S., Bergstrom, J.B., Lindskog, E., and Sridhar, T., “Computational Modeling of Dough Sheeting and Physical Interpretation of the Non-linear Rheological Behavior of Wheat Flour Dough,” Journal of Food Engineering, vol. 100, pp. 278-288, 2010.
7. Engelberg, I., and Kohn, J.,“Physico-mechanical Properties of Degradable Polymers used in Medical Applications: A Comparative Study,” Biomaterials, vol. 12, pp. 292-304, 1991.
8. Garlotta, D., “A Literature Review of Poly(Lactic Acid),” Journal of Polymers and the Environment, vol. 9, pp. 63-84, 2001.
9. Grabow, N., Schlun, M., Sternberg, K., Hakansson, N., Kramer, S., and Schmitz, K., “Mechanical Properties of Laser-Cut Poly(L-Lactide) Micro-Specimens: Implications for Stent Design, Manufacture, and Sterilization,” Journal of Biomechanical Engineering, vol. 127, pp. 25-31, 2005.
10. Ikada, Y., and Tsuji, H., “Biodegradable Polymers for Medical and Ecological Applications,” Macromolecular Rapid Communications, vol. 21, pp. 117-132, 2000.
11. Nelder, J.A., and Mead, R., “A Simplex Method for Function Minimization”, Computer Journal, pp. 308-313, 1965.
12. Perego, G., Cella, G.D., and Bastioli, C., “Effect of Molecular Weight and Crystallinity on Poly(lactic acid) Mechanical Properties,” Journal of Applied Polymer Science, vol. 59, pp. 37-43, 1996.
13. Xiao, X., “On the Measurement of True Fracture Strain of Thermoplastic Materials,” Polymer Testing, vol. 27, pp. 284-295, 2008.

8. Acknowledgement

The authors thank the BVS team at Abbott Vascular for their technical support. Thanks to Savuth Vann and Nora Legarda for their assistance with the testing.