

A Comparative Study of the Linear-elastic and Hyperelastic Models for Degradation of PLA Prepared using Fused Filament Fabrication

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FFF 방식으로 제작된 PLA의 열화에 따른 선형탄성 및 초탄성 모델의 비교에 관한 연구

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Abstract

Fused filament fabrication (FFF) is a process extruding and stacking materials. PLA materials are one of the most frequently used materials for FFF method of 3D printing. Polylactic acid (PLA)-based materials are among the most widely used materials for FFF-based three-dimensional (3D) printing. PLA is an eco-friendly material made using starch extracted from corn, as opposed to plastic made using conventional petroleum resin; PLA-based materials are used in various fields, such as packaging, aerospace, and medicines. However, it is important to analyze the mechanical properties of these materials, such as elastic strength, before using them as structural materials. In this study, the reliability of PLA-based materials is assessed through an analysis of the changes in the linear elasticity of these materials under thermal degradation by applying a hyperelastic analytical model.

Keywords : : Fused Filament Fabrication(FFF), Polylactic acid(PLA), Linear Elastic Model(선형탄성모델), Hyper Elastic Model(초탄성모델), Thermal Degradation(열화)

1. Introduction

Three-dimensional (3D) printing is currently attracting considerable attention^[1]. Compared with the existing processes such as cutting and grinding, it is

possible to manufacture complex structures easily and rapidly by using 3D printing, which has a relatively straight forward process. Moreover, 3D printing reduces process duration and cost substantially. According to previous studies, in the construction industry, 3D printing can reduce construction waste by 30-50%, labor costs by 50-80%, and production time by 50-70%^[2]. By leveraging these advantages, the

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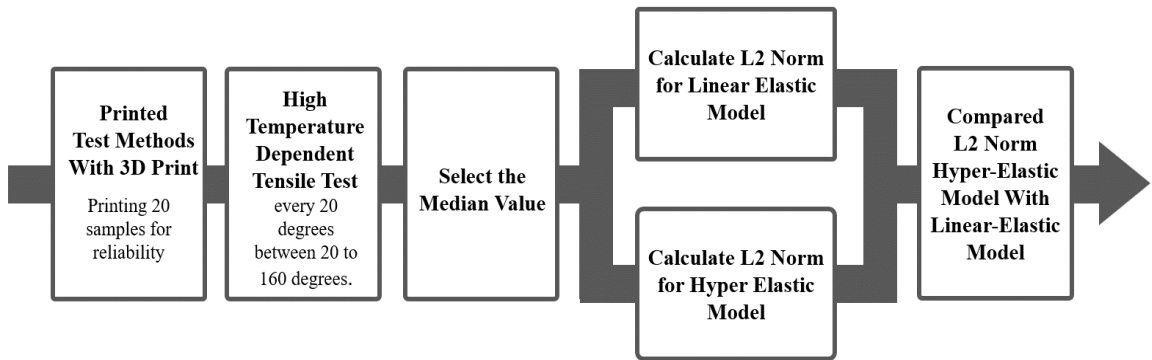


Fig. 1 Processing

company has made progress in diverse industries, including aerospace, machinery, and medicines.

NASA, for example, is working on a 3D printer for manufacturing wrenches, and for recycling a printed output to 3D print a new structure of a rocket^[3-5]. In the medical field, 3D printers are being used to produce organs for transplantation or to print custom devices that help individuals with bone regeneration^[6-7].

3D printing is performed using various methods such as fused filament fabrication (FFF) extrusion, jet, photopolymerization, powder sintering, stretching, and sheet lamination. FFF, better known as fused deposition modeling (FDM) and classed as an extrusion method, was patented in 1989 by Scott Crump, a co-founder of the Stratasys Company^[8]. Because the trademark FDM is owned by Stratasys Company, RapRap uses FFF for legal purposes. Because the patent rights of FDM expired in October 2009, various industries are now contemplating using the method^[9].

Fig. 1 briefly introduces the process of the FFF method. Thermoplastic material is filamentized and passed through a hot head, where it is heated to temperatures close to its melting point. The extruded filaments are stacked one by one in a layer-by-layer manner with repeated lamination and hardening to print a 3D structure^[8,10]. Compared to other 3D

printing methods, FFF is simple to use and inexpensive, and it requires no additional post-processing after printing. Therefore, this method can be easily used even at home or by beginners who do not have any expertise in 3D printing.

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Various materials such as polymers, metals, and ceramics, are processed using FFF. Among these materials, PLA is often used in conjunction with acrylonitrile butadiene styrene. PLA is a bioplastic made from corn starch; it is non-toxic, non-irritating and biocompatible^[12]. Moreover, it degrades completely in landfills over a period of several months to years. Therefore, PLA-based materials are attracting attention as substitutes for artificial synthetic plastic resins at a time when awareness about environmental pollution, such as the problems caused by waste disposal is increasing.

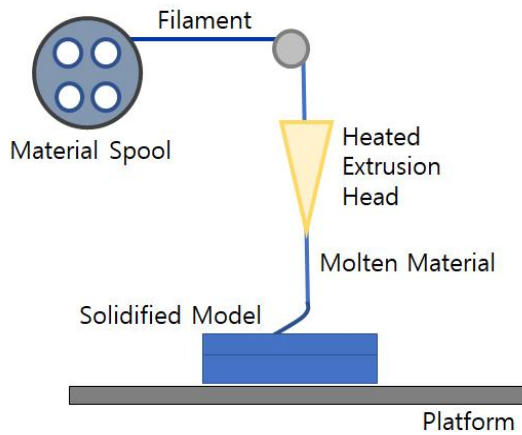


Fig. 2 Processing of FFF method

Moreover, PLA has a low melting point of around 170°C, which makes it easy and cost-effective to use^[13].

To expand the range of application of 3D printing technology and PLA-based materials, it is important to analyze the mechanical properties such as elastic modulus and reliability of PLA-based materials.

The application of FFF technology to PLA-based materials has been studied in terms of the effects of parameters on tensile strength, the effect of filament color on physical properties, and changes in mechanical properties according to the 3D printing angle^[10, 14-15].

PLA-based materials are vulnerable at high temperatures, and changes in their physical properties around at 80°C warrant extensive research on the thermal degradation phenomena of these materials^[16]. The present study aims to evaluate the linear-elastic and hyper-elastic models of thermal degradation of 3D printed PLA specimens.

2. Methodology

2.1 3D-printed Specimen

To evaluate the elastic modulus of a PLA

specimen fabricated using a FFF-type 3D printers as a function of thermal degradation, a test piece was fabricated and subjected to a tensile test. Tensile specimens of PLA were printed using a Sindoh DP-201 3D printer according to the ASTM 638-4 specifications. The nozzle temperature, bed temperature, printing speed, lamination thickness, and specimen internal density were set to 200°C, 60°C, 40 mm/s, 0.2 mm, and 100% respectively.

2.2 High-temperature Storage Test and Tensile Test

A high-temperature storage test was conducted to evaluate the changes in the properties of the specimens' material with thermal degradation. By using the ES PEC SH-662 Bench-top Type Temp (& Humi) Chamber model, 20 specimens were subjected to temperature changes in eight equal steps of 20°C from 20°C to 160°C. The maximum temperature of 165 ~ 175°C of PLA material was designated as the lower temperature of 160°C. Each specimen was subjected to the thermal degradation test for 24h. Tensile tests were conducted to evaluate the mechanical properties of the specimens by using a universal testing machine (KDMT-156). The UTM crosshead speed was set to 0.3mm/min.

2.3 Median Value

To quantitatively analyze and compare the linear-elastic and hyperelastic models, the middle value of the 20 specimens by temperature was taken as the representative value.

The average of two medians of yield strengths of the 20 specimens was computed, and the strain values were divided into 10 equal intervals. Thereafter, a test piece with the highest median value among the stress values opposed to the strains of the 20 test pieces was selected. The table in Fig. 3 lists the median values of the 20 specimens fabricated using a printing angle of 20°.

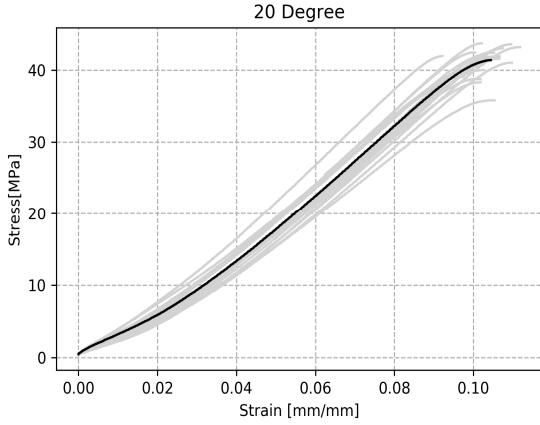


Fig. 3 Median value

2.4 Linear elastic model

The linear modulus of elasticity is based on Hooke's law and is called Young's modulus or elastic modulus, Hooke's law defines the relationship between stress and strain as follows.

$$\sigma = E \cdot \epsilon \quad (1)$$

E is the elastic modulus, σ the stress, ϵ the strain. The modulus of elasticity indicates the stiffness of a material. The higher the elastic modulus of a material, the more difficult it is to deform the material. Stress and strain increase proportionally and the stress-strain curve is straight with a certain threshold.

2.5 Yeoh Model

The Yeoh model is a nonlinear hyper-elastic model, in which the stress-strain curve is not linear. In elastic deformation, stress is proportional to strain. By contrast, a material that recovers its initial shape when the load is removed, even when the object is deformed by approximately 500% or more, is called a hyper-elastic material. Generally, rubber-based materials exhibit hyper-elasticity. Several hyper-elastic models have been developed, such as Mooney-Rivlin, Ogden, and Neo-Hooke models. In this study, we employed the Yeoh model.

$$W = \sum_{i=1}^n C_i (I_1 - 3)^i \quad (2)$$

The Yeoh Model can accurately predicting the behavior of elastomers in uniaxial tension tests and shear tests with large variations in deformation compared to Mooney-Rivlin or the Neo-Hooke models^[19]. The models for evaluating hyper-elastic materials could be used in finite element software packages such as ANSYS, and ABAQUS.

2.6 L2 Norm

The L2 norm, also called the Least Squares Error, minimizes the sum of squares of the absolute differences between the target and the estimated values.

$$s = \sqrt{\sum_{n=1}^i (y_i - f(x_i))^2} \quad (3)$$

where y_i is the target value, and x_i is the estimated value. The L2 norm was used in this study to evaluate whether the linear or the hyper-elastic model yielded a superior approximation of stress-strain curves.

3. Results and Discussion

3.1 Evaluation of Material Models According to Thermal Degradation

In this study, changes in the properties of PLA due to thermal degradation were observed in steps of 20°C over the temperature range of 20-160°C. Table 1 and Fig. 4 summarize and show, respectively, the changes in yield strength, strain, and elastic modulus with temperature. Yield strength remains constant at approximately 41MPa and then decreases rapidly to 15.87MPa at temperatures equal to or higher than 160°C. Strain remains unchanged at 0.1mm / mm until 40°C, increases from 60°C and eventually decreases to 0.07 at 160°C. Modulus of elasticity does not change considerably from its initial value of 400MPa until 4

0°C in the thermal degradation test, but decreases to 300MPa and, eventually, to 230MPa at 160°C.

In Fig. 4, similar behaviors are shown for each group when 20 and 40 degrees are designated as 1 group, 60 to 140 degrees are designated as 2 groups, and 160 degrees are designated as 3 groups. An ANOVA analysis of the E values, of the specimens yielded a P value as low as 0.00036 and an F ratio of 57.4, which is higher than the F rejected value of 5.79. Statistically, the three groups can be considered to exhibit similar behaviors.

3.2 L2 Norm Rating

The L2 norm was used to evaluate the linear-elastic and hyper-elastic models that yielded results closer to the measured values for identifying the most suitable model. Fig. 6 shows a schematic diagram of the process. For each measured value, the strain value associated with the highest yield was

Table 1 Thermal degradation of yield stress, strain, elastic modulus

Temperature	Yield Strength [MPa]	Strain [mm/mm]	Elastic Modulus
20	41.27±1.94	0.10±0.004	397.73±22.43
40	42.93±1.32	0.11±0.004	406.44±20.06
60	40.87±1.39	0.15±0.01	279.78±15.96
80	44.65±1.00	0.14±0.01	317.76±21.31
100	41.94±1.06	0.13±0.01	317.52±23.47
120	41.67±1.70	0.14±0.02	298.56±31.30
140	40.20±2.31	0.13±0.01	308.91±26.78
160	15.88±2.87	0.07±0.01	232.55±29.50

Table 2 Norm L2 comparing linear elastic to hyper elastic

	20°C	40°C	60°C	80°C	100°C	120°C	140°C	160°C
L2 Norm for Linear Elastic	19.66	8.86	14.92	20.46	13.61	21.12	20.99	5.70
L2 Norm for Hyper Elastic	5.22	7.21	6.09	6.26	5.62	5.54	4.42	2.91

divided into 10 equal intervals to obtain the corresponding stress value. Table 2 lists the calculated L2 norm values.

The yield stress, which is the highest stress, has the same linear coefficient as the measured stress-strain curve, and therefore, the final value of the L2 norm is zero. Nevertheless, the result confirmed

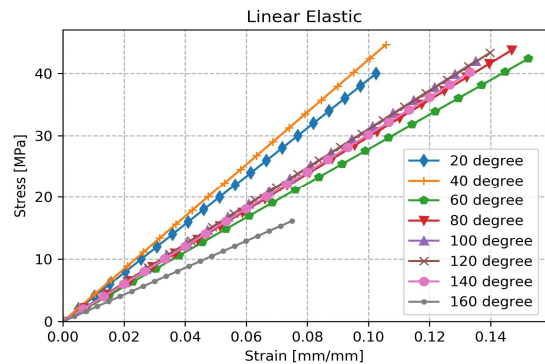


Fig. 4 Thermal degradation of yield stress, strain, elastic modulus

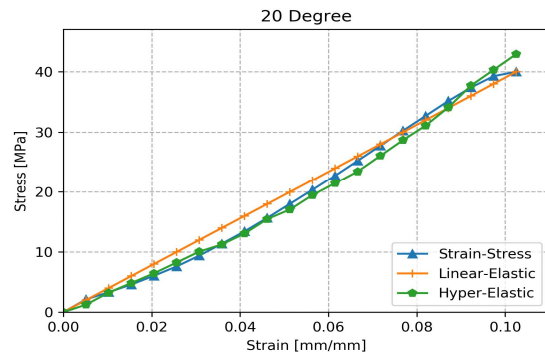


Fig. 5 Shows the graphs of the measured stress-strain curves, linear and hyper-elastic models at 20°

that L2 norm value of the hyper-elastic model was lower than that of the linear elastic model at all thermal degradation temperatures from 20°C to 160°C.

Therefore, the Yeom model, which is a hyper-elastic model, was considerably closer to the actual stress-strain curve than the linear-elastic model, which implies that the former is more suitable for our purpose.

4. Conclusion

In this study, we observed the changes in the physical properties of PLA-based materials, such as yield strength, strain, and elastic modulus, as a function of the thermal degradation temperature to evaluate their reliability. The L2 norm was used to identify the closeness of the results obtained using the linear-elastic model and the hyper-elastic model, also called the Yeom model, to the measured data. The conclusions of this study are as follows.

1. The physical properties of PLA changed as it underwent thermal degradation. The strain slightly increased between 60°C and 140°C and then rapidly decreased at 160°C. The modulus of elasticity rapidly decreased at thermal degradation temperatures of 60°C and higher. The yield strength of the material did not significantly change at thermal degradation temperatures of up to 140°C, and it significantly decreased from 41MPa to 15MPa at 160°C.
2. A comparison of Hooke's linear-elastic model and the Yeoh hyper-elastic model using in terms of the L2 norm; values revealed that the L2 norm value of the Yeoh model was lower L2 than that of the linear-elastic model.

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REFERENCES

1. Shahrubudin, N., Lee, T. C., Ramlan, R., "An Overview on 3D Printing Technology: Technological, Materials, and Applications", *Procedia Manufacturing*, Vol. 35, pp. 1286-1296, 2019.
2. Doris(2016), "3D Concrete Printing Market to Reach \$ 56.4 Million by 2021", Retrieved, 31, Dec., 2019, from <https://www.3pnr.com/3d-concrete-printing-market-reach-56-4-million-2021-1239664/>
3. Wall, M. (2014), "Space Station's 3D Printer Makes Wrench From 'Beamed Up' Design", Retrieved, 31, Dec., from <https://www.space.com/28095-3d-printer-space-station-ratchet-wrench.html>
4. Anderson, J. (2017), "Full Circle: NASA to Demonstrate Refabricator to Recycle, Reuse, Repeat", Retrieved, 31, Dec., from https://www.nasa.gov/mission_pages/centers/marshall/images/refabricator.html
5. Urrutia, D. E. (2019), "Relativity Space to Launch Satellite 'Tugs' on 3D-Printed Rocket", Retrieved, 31, Dec., from <https://www.space.com/relativity-space-to-launch-momentum-tugs-2021.html>
6. Yana, Q., Dong, H., Sua, J., Han, J., Song, B., Wei, Q., Shia, Y., "A Review of 3D Printing Technology for Medical Applications", *Engineering*, Vol. 4, No. 5, pp. 729-742, 2018.
7. Hassan, M. N., Yassin, M. A., Suliman, Lie, S., S. A., Gjengedal, H., Mustafa, K., "The bone regeneration capacity of 3D-printed templates in calvarial defect models: A systematic review and meta-analysis", *Acta Biomaterialia*, Vol. 91, pp. 1-23, 2019
8. Bryll1a, K., Piesowicz, E., Szymański, P.,

- Ślącza, W., Pijanowski, M., "Polymer Composite Manufacturing by FDM 3D Printing Technology", MATEC Web of Conferences, Vol. 237, 2018.
9. Seol, K. S., Shin, B. C., Zhang, S. U., "Fatigue Test of 3D-printed ABS Parts Fabricated by Fused Deposition Modeling", Journal of the Korean Society of Manufacturing Process Engineers, Vol. 17, No. 3, 2018.
10. Lee, S. K., Kim, Y. R., S. Kim, H., Kim, J. H., "Investigation of the Internal Stress Relaxation in FDM 3D Printing : Annealing Conditions", Journal of the Korean Society of Manufacturing Process Engineers, Vol. 17, No. 4, pp. 130-136, 2018.
11. Farah, S., Anderson, D. G., Langer, R., "Physical and mechanical properties of PLA, and their functions in widespread applications — A comprehensive review", Advanced Drug Delivery Reviews, Vol. 107, pp. 367-392, 2016.
12. You, Y. S., Oh, Y. S., Hong, S. H., Choi, S. W., "International Trends in Development, Commercialization and Market of Bio-Plastics", CLEAN TECHNOLOGY, Vol. 21, No. 3, pp. 141-152, 2015.
13. Jang, M. O., Nam, B. U., Jeong, D. S., Hong, C. H., "Characteristics of Preparation and Thermal Properties of PLA Stereocomplex", Prospectives of Industrial Chemistry, Vol. 14, No. 1, pp. 1-4, 2010.
14. Rao, MV. D. P., Rajiva, P., Geethika, V. N., "Effect of fused deposition modelling (FDM) process parameters on tensile strength of carbon fibre PLA", Materials and Design, Vol. 14, No. 18, No. 6, pp. 2012-2018, 2019.
15. Wittbrodt, B., Pearceab, J. M., "The effects of PLA color on material properties of 3-D printed components", Additive Manufacturing, Vol. 8, pp. 110-116, 2015.
16. Choi, W., Woo, J. H., Jeon, J. B., Yoon, S. S., "Measurement of Structural Properties of PLA Filament as a Supplier of 3D Printer", Journal of the Korean Society of Agricultural Engineers, Vol. 57, No. 6, pp. 141-152, 2015.
17. Gajewski, M., Szczerba, R., Jemiolo, S., "Modelling of Elastomeric Bearings with Application of Yeoh Hyperelastic Material Model", Prospectives of Industrial Chemistry, Vol. 111, pp. 220-227, 2015.