

ADDITIVE MANUFACTURING OF MULTILAYERED POLYMER COMPOSITES: DURABILITY ASSESSMENT

Ali, Payami Golhin^a, Chaman, Srivastava^a, Jens Fossan, Tingstad^b, Aditya Suneel, Sole^b, Are, Strandlie^a, Sotirios, Grammatikos^a

a: Laboratory for Advanced and Sustainable Engineering Materials (ASEMlab), Department of Manufacturing and Civil Engineering, Norwegian University of Science and Technology, 2815 Gjøvik, Norway – ali.p.golhin@ntnu.no

b: Department of Computer Science, Norwegian University of Science and Technology, 2815 Gjøvik, Norway

Abstract: *This study examines the impact of additive manufacturing (AM) settings on the durability of 3D-printed polymer composite objects in terms of object color appearance and corresponding mechanical properties when subjected to simulated environmental conditions using accelerated aging. For this purpose, the AM pre-processing factors that influence the performance of the composite material, such as build platform position, color, and finishing configurations, are discussed. The experimental campaign was designed according to the Taguchi method to minimize the color difference and maximize the mechanical parameters. The results indicate that the best factor parameters for each performance characteristic differ following the design goal. Accordingly, black-on-white with a glossy-on-matte finish manufactured on the outer swath demonstrated the best color and mechanical performance fidelity, where the studied properties were not altered significantly due to aging.*

Keywords: PolyJet 3D printing, Accelerated aging, Structural degradation, Object appearance, Color change

1. Introduction

The new r-theta rotating build platform approach with a fixed print head was recently introduced as an alternative to traditional XYZ build platform 3D printers. The stated reasons for the disc-shaped build platform include increased reliability, easier maintenance, and a smaller footprint. However, the distinct layering mechanism and pattern compared to other AM machines necessitates new considerations in the design, appearance, and durability evaluation. In particular, a close inspection of the various assembled models printed on a rotary build platform reveals a few broken and glued small parts, indicating that the commercial PolyJet printer currently in use produces rigid and brittle parts with microscopic textures [1, 2]. In addition, while Material jetting (MJT)-printed products represent a desirable appearance, their durability is vital due to their prohibitive costs and environmental impact, especially for engineering applications [3].

The surface quality of 3D-printed objects can be affected by various process parameters. According to the Taguchi approach in the design of experiments, the process variables can be reduced, and the mean values must be adjusted to a target value. Orthogonal arrays are used to execute the tests and measure process quality. Thus, the signal-to-noise (S/N) ratios can

assess the quality of the production process designed for regular application in testing, surgical planning, and tooling [4].

In this work, we present the results of an investigation into the optical and mechanical properties of 3D-printed bi-layer polymer specimens manufactured under various parametric conditions. The effect of three 3D printing parameters, including color, the position of the piece on the build platform (swath selection), and finishing on the mechanical and appearance variables, was investigated using the Taguchi methodology. The novelty of the work presented herein lies in the unique linkage between appearance and long-term mechanical properties, providing new insight into assessing the durability of AM products.

2. Materials and methods

A Stratasys J55 PolyJet 3D printer has been used to manufacture samples under different conditions. The characteristics of the VeroPureWhite (RGD837), VeroBlackPlus (RGD875), VeroCyan (RGD843), VeroYellow (RGD836), and VeroMagenta (RGD851) photo-resins (Stratasys Ltd., USA) are investigated in this study. These rigid polymers mainly consist low-viscosity acrylic oligomer, *exo*-1,7,7-trimethylbicyclo [2,2,1] hept-2-yl acrylate, (octahydro-4,7methano-1H-indenediyl) bis(methylene) diacrylate, and 4-(1-oxo-propenyl)-morpholine [5].

Parts were designed with a minimum thickness of 2 mm when a 1 mm white background was covered by 1 mm of colored material. The reference materials for color studies were the same as-printed samples before aging. For mechanical testings, the specimens were selected from the magenta samples printed on the middle swath and glossy on the glossy (GoG) finish. According to the datasheet for used photo-resins [6], the Vero materials family shares similar mechanical, thermal, and electrical properties.

To investigate the impact of 3D printing parameters on the variables, an L8(4¹ 2²) orthogonal array was chosen for three parameters of color, swath, and finishing process. As a result, 63 specimens were manufactured for 8 runs and as references. The process parameters and their levels are summarized in Table 1.

Table 1: Taguchi factors and their levels.

Run	Factor		
	Color	Swath	Finish
1	Cyan	Inner	Glossy on Glossy
2	Cyan	Outer	Glossy on Matte
3	Magenta	Inner	Glossy on Glossy
4	Magenta	Outer	Glossy on Matte
5	Yellow	Inner	Glossy on Matte
6	Yellow	Outer	Glossy on Glossy
7	Black	Inner	Glossy on Matte
8	Black	Outer	Glossy on Glossy

For the assessment of mechanical performance, tensile and viscoelastic testing were considered (Figure 1).

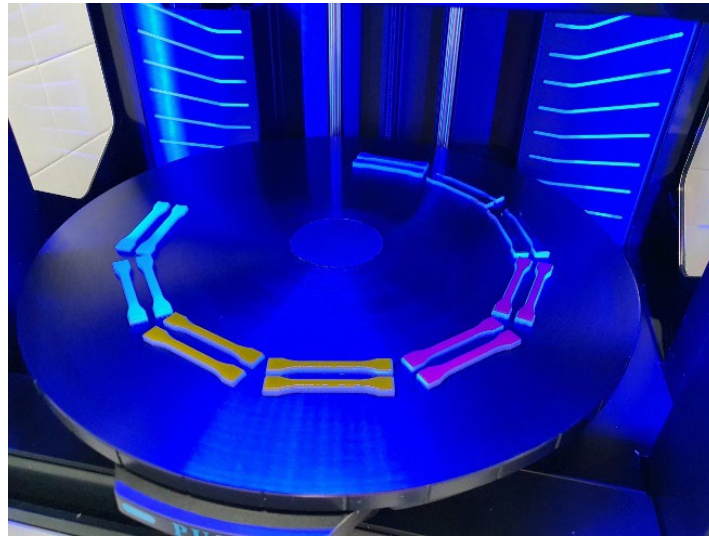


Figure 1. Reference tensile specimen in the middle swath of the rotary build platform.

To determine the durability of the polymeric specimens, the specimens were aged in a QUV chamber from QLab for a maximum duration of 2 weeks. The aging regime representing natural weathering was selected according to the ASTM G154 standard. The weathering cycle had a UV exposure at 0.8 W/m^2 for 6 hours at $60 \text{ }^\circ\text{C}$, followed by condensation at $40 \text{ }^\circ\text{C}$ and spray at $24 \text{ }^\circ\text{C}$, where the humidity is set at 60% and 100%, respectively.

A Konica Minolta CS-2000 tele-spectroradiometer (TSR) was used to measure appearance by recording radiance at the specimen surface within the 380-780 nm spectral range. The optical resolution and physical sampling intervals were 1 and 10 nm, respectively. The device was placed 50 cm in front of the target object, normal to the surface (Figure 2). The measurement field of view was set to 0.2 degrees to avoid error due to targeting areas covered by stains or dust. The surface of the 3D-printed specimen was studied using a $45^\circ:0^\circ$ viewing geometry according to CIE Publication 15.2 [7]. The spectral results of targets were calibrated using the standard spectralon white ceramic patch. The colorimetric values were calculated using the computational color science toolbox in MATLAB R2021a [8].

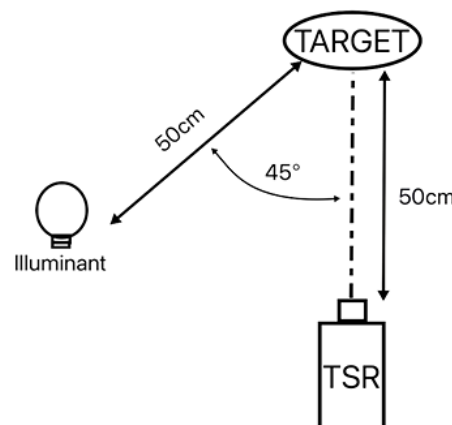


Figure 2. Schematic view of the color appearance measurement setup.

Dynamic Mechanical Analysis (DMA) of the polymer coupons is carried out to determine the glass transition temperature (T_g) and the relative shift in the T_g as a function of the aging time and temperature [9]. The test was performed on Discovery DMA 850 from TA instruments, as per the ASTM D4065 standard. The specimen dimensions were $60 \times 13 \times 3 \text{ mm}^3$. A temperature ramp test was performed at a heating rate of $2 \text{ }^\circ\text{C}$. The test temperature range was set from 20°C to 95°C , which was below the degradation temperature of the material. The amplitude of the test was set to $20 \text{ }\mu\text{m}$ with a frequency of 1 Hz . The maxima of the $\tan \delta$ curve and its corresponding temperature are referred to as the T_g reported in the paper.

To determine the tensile modulus and the strength of the polymer material as a function of aging duration, ISO 527-02 was used [10]. The dimensions of the dog bone specimens were $75 \times 10 \times 2 \text{ mm}^3$. Uniaxial tensile testing was performed on Instron 5966 universal testing machine, and the strain was measured using an AVE2 virtual extensometer. The loading rate was set to 2 mm/min . The force-displacement curves were used to determine the tensile chord modulus, and the ultimate tensile stress was chosen as the tensile strength of the bi-layer material.

3. Results and discussions

Figure 3 depicts the color difference for DMA coupons after two weeks of accelerated weathering. According to CIEDE2000 colorimetric difference [7], in Figure 3a, magenta samples are notably altered in color compared to reference samples. On the other hand, cyan and yellow samples represent noticeable color differences compared to the control parts and at a lower level. Similar advantages for cyan color over red feedstock were described before for fused deposition modeling (FDM) [11]. In magenta and yellow samples, significantly higher color differences were observed for the GoG samples.

On the other hand, cyan and black objects respond in reverse as their average CIEDE2000 has slightly decreased. The redness (a^*) and yellowness (b^*) shifts in the CIE $L^*a^*b^*$ values at a constant lightness of 90 suggest yellower yellow samples, less red magenta samples, and bluer/greener cyan samples after two weeks of aging (Figure 3b). The color of the black objects did not change enough to be recognized without measurement.

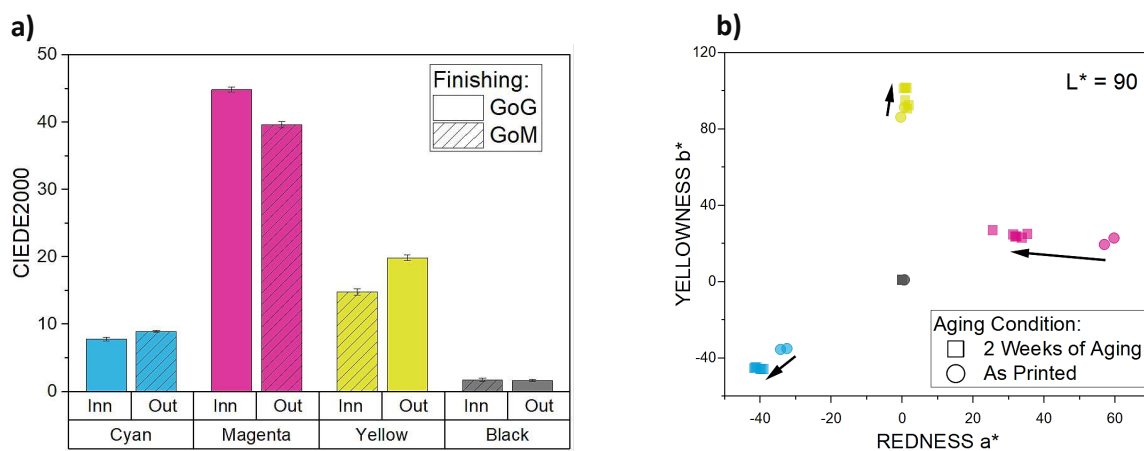


Figure 3. a) CIEDE2000 color difference and associated root mean square error (RMSE) bars, and b) shift in the a^*b^* values after the aging process.

The DMA coupons after two weeks of weathering are depicted in Figure 4. In magenta samples, the marginal dark-red area represents the original color obscured by the sample holders. As seen from the edges, the color differences for magenta samples were significant, while the color differences for cyan and yellow pieces were barely noticeable after aging.



Figure 4. DMA specimens after two weeks of weathering exposure.

Figure 5a depicts representative stress-strain curves for each type of specimen aged and tested. The results are compared with the reference specimens before aging. It is observed that the tensile modulus of the polymer specimen increases during the 2 weeks aging regime, which complements the findings from the DMA analysis and in conjunction with the stiffness increase. The table shown in Figure 5b populates the results for glass transition temperature measurements from the $\tan \delta$ curve from the DMA analysis. It is observed that the glass transition temperature of the polymeric material has increased significantly to a range of 80-84°C for each specimen, where the reference value of 53°C is taken from the reference manual for the feedstock material. The sharp increase in the change in the T_g of the material is very anomalous. This can be attributed to the material being cured during 3D printing, pre-aging due to storage, and weathering process, where the T_g increased slowly.

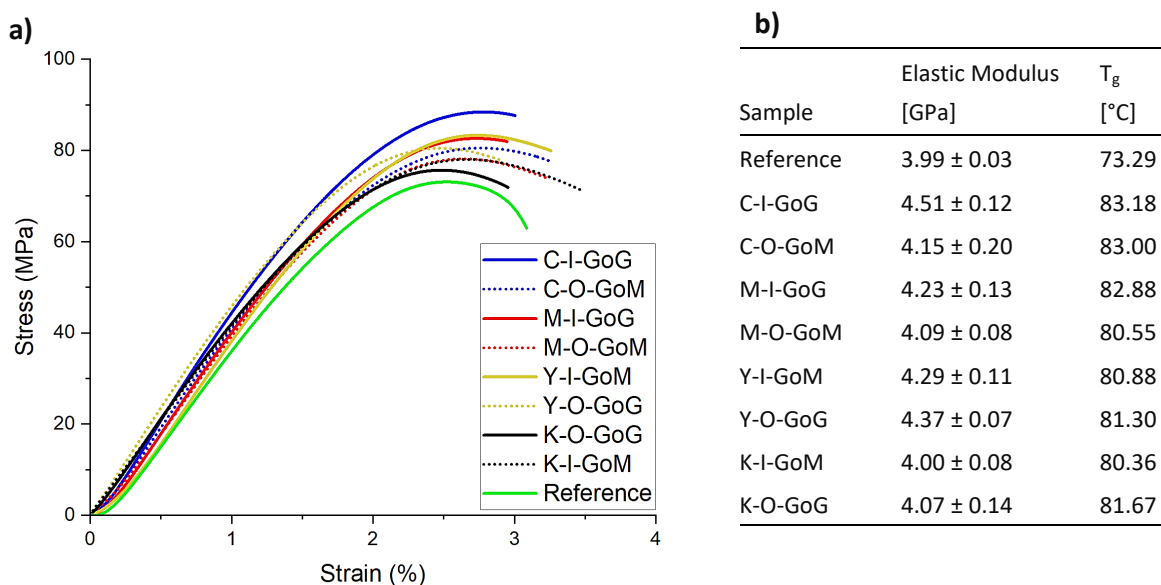


Figure 5. a) Stress-strain curves for tensile specimens, and b) E modulus and T_g results; C: Cyan, M: Magenta, Y: Yellow, K: Black, I: Inner Swath, O: Outer Swath, GoG: Glossy on Glossy finish, GoM: Glossy on Matte finish.

According to the S/N ratio plots in Figure 6a, color, as expected, had the most significant influence on the color difference. In contrast, swath and finishing had no considerable impact on this variable. However, after two weeks of weathering, the choice of outer swath and GoG finish resulted in more stable color difference calculations (Figure 6 b)). In terms of color, black and then cyan samples demonstrated significantly more reliable color fidelity than magenta and yellow samples after aging.

On the other hand, S/N ratio plots for mechanical properties (Figures 6c and d) revealed, in addition to color, that the swath and finishing significantly influenced the elastic modulus and T_g of the studied samples. Accordingly, the inner swath and GoG finish resulted in higher elastic modulus and T_g . Cyan provided higher E and T_g than black, while magenta and yellow samples demonstrated unstable behavior in mechanical testing. The ultraviolet (UV) absorbance of samples during the weathering test can explain the possible effect of post-curing behind these mechanical behaviors of the studied specimens, resulting in more rigid materials with altered mechanical properties and color appearances.

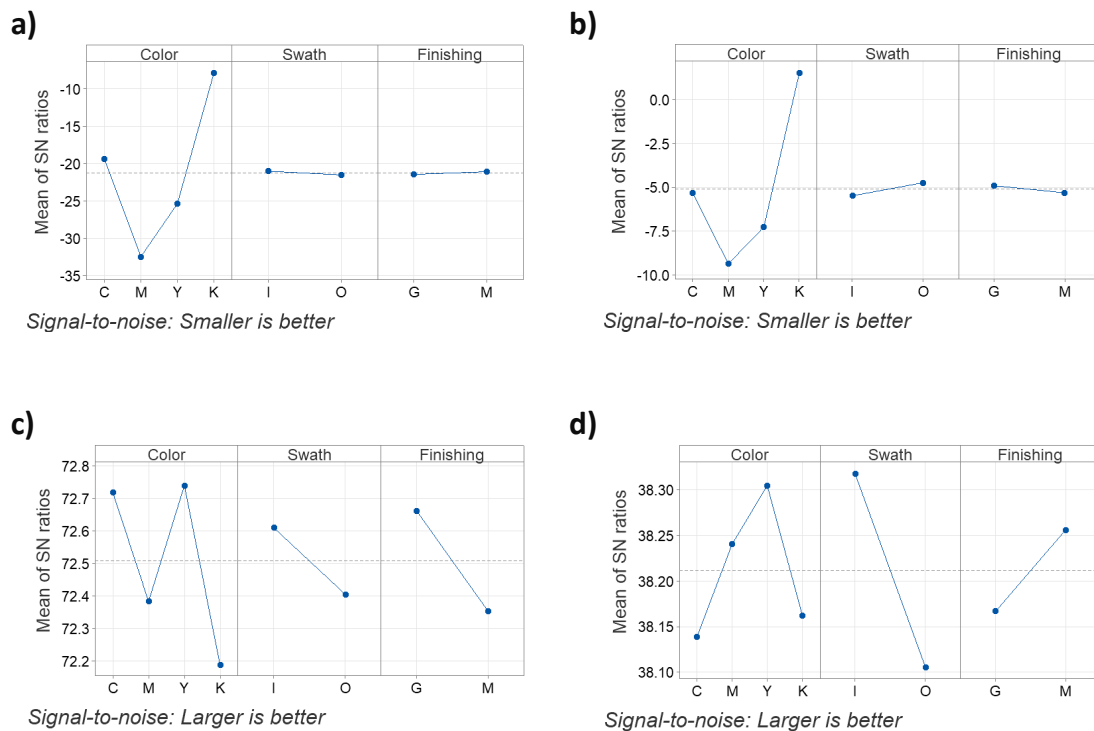


Figure 6. Main effects S/N plots for a) CIEDE2000, b) RMSE for CIEDE2000, c) Elastic modulus, and d) T_g .

4. Conclusions

A durability study was conducted to investigate the effect of processing parameters, including color, the position of the piece on the build platform (swath), and finishing on the long-term mechanical properties and appearance of photo resin samples made using the Stratasys Vero family after two weeks of accelerated aging in a weathering chamber. Based on the results, the color factor solely dominated the color appearance results compared to other studied parameters. On the other hand, the swath and finishing selections, in addition to color, also significantly affected the mechanical properties.

Compared to as-printed control samples, all aged coupons showed altered mechanical properties. However, the black-on-white with a glossy-on-matte finish manufactured on the outer swath offered the best color fidelity and stable mechanical behavior after weathering. In contrast, the magenta samples had the weakest durability response due to the possible excessive UV post-curing in the QUV chamber. Furthermore, the durability assessment in a harsh environment for yellow and cyan samples revealed their unstable behavior due to their color appearance, which should be considered in the design process. The results demonstrated the importance of the color study as a primary parameter when investigating weathering and UV exposure, which is necessary for engineering applications.

5. Future studies

The nature of the weathering test is to age the samples over an extended period. Therefore, further research on the response of variables over a more extended aging period allows for a better balance of all studied parameters to be discovered. To this end, our next step is to conduct a long-term study on the samples to reduce the impact of measurement uncertainty and provide more details to improve the durability of an object in MJT additive manufacturing.

Acknowledgments

We would like to acknowledge support from the ApPEARS-ITN project funded by the European Union's H2020 research and innovation program under the Marie Skłodowska-Curie grant agreement No. 814158 and from the LIFETIME project, Ref: 309943, funded by the Research Council of Norway.

6. References

1. Elber G, Kim MS. Synthesis of 3D jigsaw puzzles over freeform 2-manifolds. *Comput Graphics (Pergamon)*. 2021.
2. Payami Golhin A. Generation of micro-and nano-textured surfaces. Brussels: European Commission; 2021.
3. Li Y, Linke BS, Voet H, Falk B, Schmitt R, Lam M. Cost, sustainability and surface roughness quality – A comprehensive analysis of products made with personal 3D printers. *CIRP J Manuf Sci Technol*. 2017;16:1-11.

4. Camposeco-Negrete C. Optimization of FDM parameters for improving part quality, productivity and sustainability of the process using Taguchi methodology and desirability approach. *Prog Addit Manuf.* 2020.
5. Tee YL, Peng C, Pille P, Leary M, Tran P. PolyJet 3D printing of composite materials: experimental and modelling approach. *Jom.* 2020;72(3):1105-17.
6. Vero: Realistic, Multi-Color Prototypes in Less Time. 2022.
7. l'Éclairage Cld. Colorimetry CIE 015:2004: Commission Internationale de l'Éclairage; 2004. p. 1-82.
8. Westland S, Ripamonti C, Cheung V. Computational colour science using MATLAB: John Wiley & Sons; 2012.
9. Das SC, Paul D, Grammatikos SA, Siddiquee MAB, Papatzani S, Koralli P, et al. Effect of stacking sequence on the performance of hybrid natural/synthetic fiber reinforced polymer composite laminates. *Compos Struct.* 2021;276.
10. Plastics - Determination of tensile properties. Part 2: Test conditions for moulding and extrusion plastics: International Organization for Standardization ISO/TC 61/SC 2 Mechanical behavior; 2012.
11. Payami Golhin A, Strandlie A, John Green P. The Influence of Wedge Angle, Feedstock Color, and Infill Density on the Color Difference of FDM Objects. *J Imaging Sci Technol.* 2021;65(5):050408-1--15.

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Proceedings of the 20th European Conference on Composite Materials

COMPOSITES MEET SUSTAINABILITY

Vol 6 – Life Cycle Assessment

Editors : Anastasios P. Vassilopoulos, Véronique Michaud

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ECCM20 - Proceedings

ISBN: 978-2-9701614-0-0

DOI: [10.5075/epfl-298799_978-2-9701614-0-0](https://doi.org/10.5075/epfl-298799_978-2-9701614-0-0)