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Influence of Layer Thickness of 3D Printed Polyamide Against Temperature

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ABSTRACT

Polyamide is one of the materials in 3D printing that can produce valuable products to meet the needs of the industry. Previous studies have proven that the layer thickness of the 3D printed material and the increase in temperature affect the mechanical and physical properties. However, only a few studies involve polyamide material as a test material, especially in analyzing the influence of the layer thickness of the printed material and the increase in temperature on the mechanical and physical properties of polyamide. Therefore, the bending properties of polyamide with different layer thicknesses at 0.1 mm, 0.2 mm and 0.3 mm and the tensile properties of the material at different temperatures at room temperature, 75˚C and 110˚C will be studied. This study will involve polyamide (PA) materials printed at three different layer heights using the Fused Deposition Modelling (FDM) process. Bending and tensile tests at different temperatures from 27˚C to 110˚C are conducted using the Instron Universal Testing Machine. The study results show that the layer height of 0.3 mm exhibits the highest flexural strength at an average rate of 11.05 MPa compared to 0.1 mm (6.7 MPa) and 0.2 mm (9.6 MPa). The tensile strength decreases when the temperature elevates, making the temperature of 110˚C have the lowest tensile value (1.591 MPa) compared to the temperature of 75˚C (1.6MPa) and 27˚C (2.1MPa). Several material characterizations such as SEM, TGA, DMA, DSC and density have been performed to study the microstructure and influence of tensile test temperature on the mechanical properties of polyamide.

Keywords: Polyamide; 3D printing; layer height; mechanical strength; temperature

INTRODUCTION

Additive Manufacturing (AM) technology has become more widespread, especially in producing artificial heart pumps, jewellery collections, 3D printed corneas, PGA rocket engines, steel bridges and other products related to the aviation and food industries (Shahrubudin et al. 2019). Fused Deposition Modelling (FDM) is one of the most popular techniques for additive manufacturing due to its versatility in feedstock materials. The procedure involves melting the material in the form of a wire cartridge and simultaneously reducing its cross-section to sub-millimetre dimensions. Due to the relatively low processing temperature and the ability to control the molten and solidified phases, this technique is the best option for processing various polymers (Guessasma et al. 2021).

FDM can create intricate designs but has limitations that need further exploration These disadvantages include high cost, limited printing for large, structured products and mass production, inferior and anisotropic mechanical properties, limited materials, and defects in product yield (Duc Ngo et al. 2018). The success of the FDM process is hugely dependent on two factors: selecting appropriate material and using the correct FDM printing parameter

configuration. In general, amorphous polymer materials are appropriate for FDM 3D printing. Nevertheless, if a semi-crystalline polymer is utilised, the material's properties should resemble those of an amorphous polymer, particularly during cold crystallisation from a liquid polymer. Polyamides (PA), for instance, are suitable for FDM processing because some polyamides have an amorphous structure, and the remainder are classified as semi-crystalline polymers with a degree of crystallinity between 20% and 32%. Consequently, molecules have a high propensity to solidify or melt rapidly when chilled or heated (Tuan Noraihan Azila Tuan Rahim et al. 2019).

The primary objective of this study is to provide a clear picture of the potential of nylon as a candidate material for FDM by relating the influence of layer thickness on flexural strength, the resulting microstructure, and the effect of elevated temperature in tensile test towards the mechanical performance PA. Polyamide is more widely used in the automotive and aerospace industries than other high-performance polymers such as Polyphenylene Sulphide (PPS), Polyether-ether-ketone (PEEK), Polyaryl-ether-ketone (PAEK), and Polyetherimide (PEI). PA requires a lower refining temperature than conventional polymers such as polypropylene (PP) and polyethene (PE), preventing the material from being utilised with the same equipment (Kondo et al. 2022).

Layer thickness can be changed and controlled to explore the mechanical properties of 3D printed materials. Vishwas, Basavaraj & Vinyas (2018) found that polyamide material with the lowest layer height of 0.1 mm had the highest tensile strength compared to 0.2 mm and 0.3 mm. This study suggests that the mechanical characteristics of PA decrease as layer height increases. However, increasing layer heights elevates residual stress and temperature and cooling cycles. This condition can weaken specimen material by distorting, cracking, and delaminating sections or causing fabrication failure (Panda et al. 2016). Besides, Grasso et al. (2018) found that higher temperatures increase stiffness, leading to failure. PA has a semi-crystalline structure comprising crystalline and amorphous polymer chains. High temperatures can phase shift or disrupt this crystal structure, lowering tensile strength. Disrupting this crystalline zone can diminish load-bearing capability and cause deformation or failure (Vozniak & Bartczak 2023).

However, limited studies highlight a relationship between layer thickness and tensile performance when evaluated at elevated temperatures. Noting that the layer thickness will contribute to voids during the printing process, resulting in poor mechanical performance, especially when applied in electronic or robotic applications. This research can serve as an excellent foundation for future advancements in 3D printing technology, and the resulting material could be used for sustainable development in a wide range of applications.

METHODOLOGY

Polyamide sample printing using the FDM method was purchased from Sigma-Aldrich (M) Sdn Bhd. This study was conducted using thermoplastic polymer filament polyamide (PA), PAHT CF15, an industrial-grade filament with a unique formulation consisting of polyamide and 15% additional micro-carbon fibres. Table 1 explains the printing parameters of the specimen.

TABLE 1. Printing parameter

Resolution	Standard
Infill	100%
Pattern orientation	0˚
Nozzle temperature	230˚C
Printing speed	50 mm/s
Layer height	0.1mm, 0.2mm, 0.3mm

Mechanical properties. Flexural strength and strain of specimens were measured using an Instron Universal Testing machine, fitted with a 30kN load cell and at a 5 mm/min crosshead speed. The specimen dimension followed ASTM D790 standards, subjected to bending load in a horizontal position with a support span of 50.8 mm. Fifteen specimens with different layer thicknesses of 0.1 mm, 0.2mm and 0.3 mm were tested, and the average value was taken. The dimensions of the flexural specimen as shown in Figure 1.

Next, to achieve the second objective, fifteen standard tensile specimens were tested using the same testing machine under three different temperatures, namely at 27˚C, 75˚C and 110˚C. The tensile test was performed on the sample by following the ASTM D 638 standard Type 1 under a crosshead speed of 5 mm/min to failure. The tests were repeated five times for each parameter to ensure statistical significance. The dimensions of the flexural specimen as shown in Figure 2.

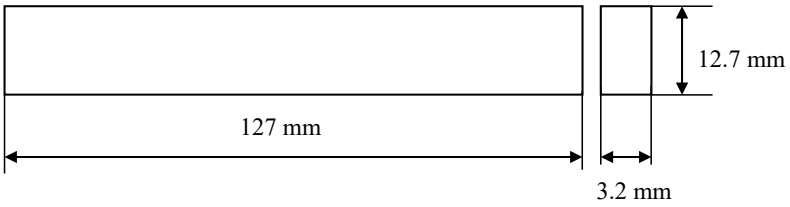


FIGURE 1. The dimension of the flexural sample

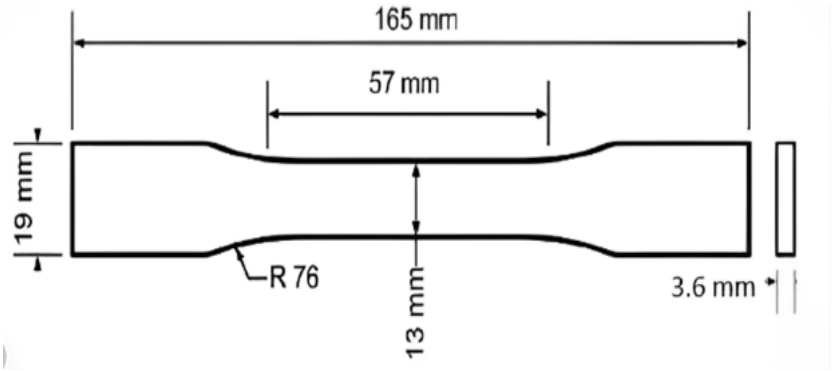


FIGURE 2. The dimension of the tensile sample

DENSITY

The density of the PA sample was measured by using Sartorius BSA224S-CW densimeter. A 10 mm x 13 mm dry sample was weighed using a densimeter and then completely immersed in distilled water at room temperature. Five measurements were carried out, and the average value was taken.

SCANNING ELECTRON MICROSCOPE (SEM)

The tests were conducted according to ASTM E 986-04 standards using a Hitachi Tabletop Microscope TM-1000 machine. SEM was performed at 60x, 80x and 100x magnifications with an accelerating voltage of 10.0 kV to obtain a cross-sectional image of the fractured polyamide microstructure.

THERMOGRAVIMETRIC ANALYSIS (TGA) AND DIFFERENTIAL SCANNING CALORIMETRY (DSC)

TGA and DSC were carried out on PA samples weighing 20 mg using a TGA DSC testing tool, Mettler Toledo TGA/DSC 1. Specimens were heated at 20˚C/min from 30˚C to 800˚C under a nitrogen atmosphere.

DYNAMIC MECHANICAL ANALYSIS (DMA)

This test was performed based on ASTM D 4065 standards using a Perkin Elmer model DMA8000 machine in dual cantilever mode. The temperature range was between 25˚C and 150˚C at a frequency of 1 Hz with a heating rate of 1˚C/min.

RESULTS AND DISCUSSION

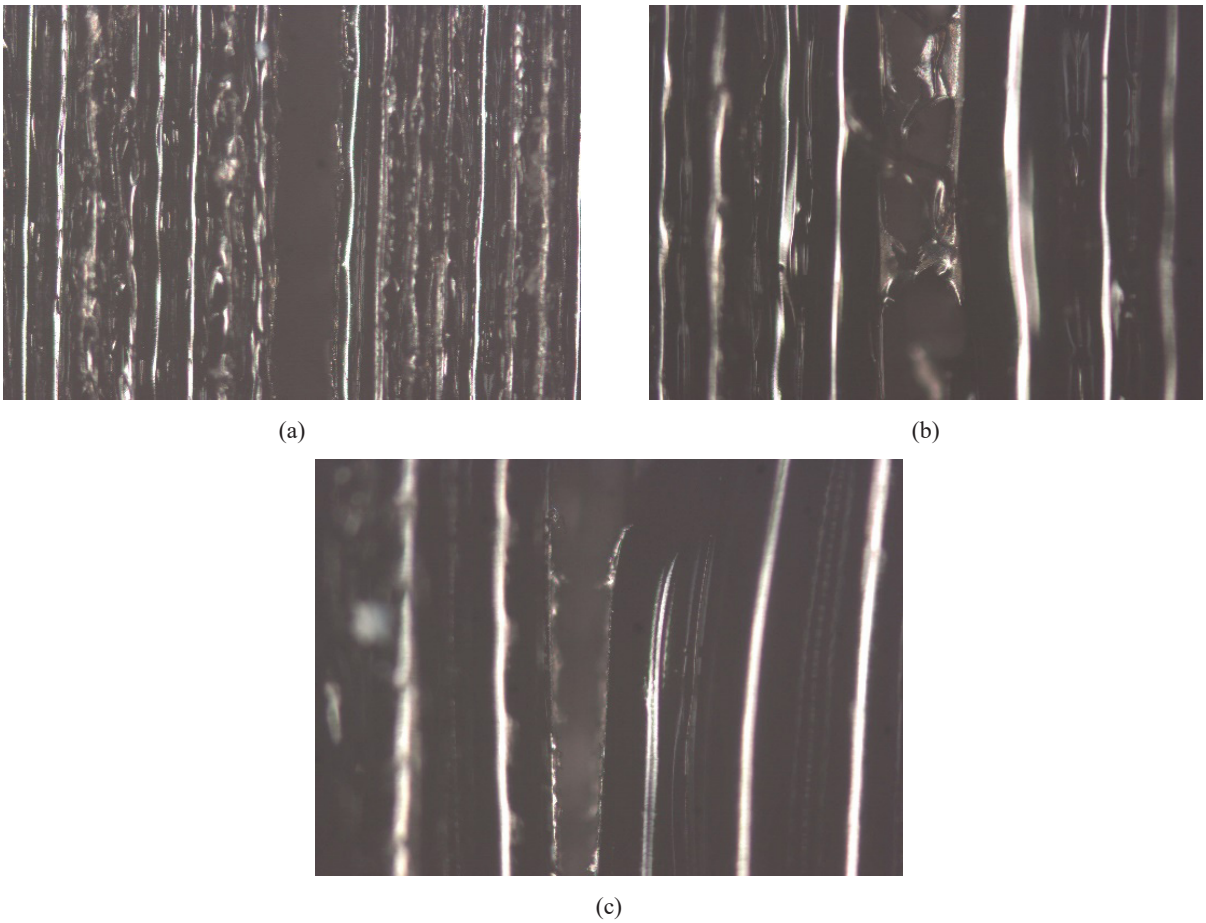
EFFECT OF LAYER HEIGHT ON POLYAMIDE BENDING STRENGTH

Table 2 shows the flexural properties of polyamide for layer thickness 0.1 mm, 0.2 mm, and 0.3 mm. The layer height of 0.3 mm shows the highest bending strength compared to 0.1 mm and 0.2 mm, with an average bending strength of 11.05 MPa. On the other hand, the layer height of 0.1 mm shows the lowest bending strength with an average rate of 6.739 MPa, followed by 0.2 mm, with a strength of 9.636 MPa. According to Panda et al. (2016), increasing the number of layers also increases the number of heating and cooling cycles resulting in increased residual stress accumulation. This situation can result in distortion, cracking between layers and delamination of parts or fabrication failure, thus reducing the strength of the specimen material.

TABLE 2. Flexural properties of polyamide

Layer height (mm)	Flexural strength (MPa)	Flexural strain (%)	Elongation (mm/mm)
0.1	6.739	2.097	2.818
0.2	9.636	12.666	17.024
0.3	11.050	12.707	17.079

There are numerous causes for residual tensions in the FDM of semi-crystalline polymers. During the cooling phase, the extruded polymer material that has been deposited cools to the ambient temperature. The outer surface cools considerably quicker than the interior. This discrepancy in the thermal profile produces compressive stress on the exterior surface. In contrast, tensile tension is formed within the polymer to counterbalance, resulting in anisotropic contraction (shrinkage), internal residual stress buildup, and warpage. The study concluded that layer thickness directly affects part distortion and built-in residual stress (Samy et al. 2021)



(a) (b) (c)

FIGURE 3. Separated layer of the bending specimen sample at layer height (a) 0.1 mm (b) 0.2 mm (c) 0.3 mm

Figure 3 shows the microstructural changes in the arrangement of nylon filaments, especially in the fractured area. When the bending load is applied to the sample parallel to the orientation alignment, the load is concentrated in the central area of the sample, which causes the failure of the specimen to occur due to the separation of the layers from the bonded area. At a layer height of 0.1 mm, the printed PA layer starts to separate, resulting in large voids in the surface area of the sample. However, the separation gap between the layers decreases as the layer thickness increases. The 0.3 mm layer has formed a smaller gap than the 0.2 mm. This SEM visual proves that the 0.3 mm layer has a stronger bond or adhesion between the layers compared to 0.1 and 0.2 mm.

EFFECT OF ELEVATED TEMPERATURE ON THE TENSILE STRENGTH OF POLYAMIDE

Table 3 exhibits the tensile test results at different test temperatures, namely at 27˚C, 75˚C and 110˚C at layer thickness 0.3 mm. The results show that increasing the temperature of the tensile test has caused the tensile

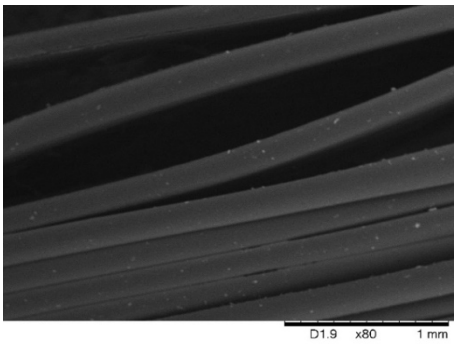
strength of PA to decrease. A temperature of 110˚C shows the lowest tensile strength, with an average rate of 1.591 MPa, compared to a temperature of 75˚C and 27˚C, with an average tension rate of 1.668 MPa and 2.159 MPa. As expected, an increase in temperature results in a higher tensile strain at failure.

TABLE 3. Tensile properties of polyamide

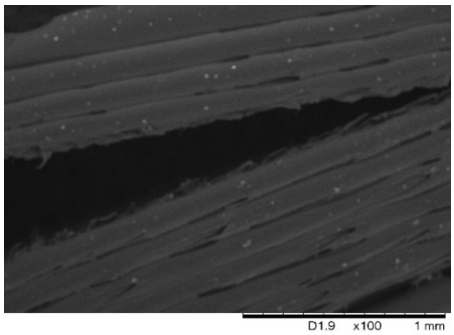
Temperature (˚C)	Tensile strength (MPa)	Tensile strain (%)	Elongation (mm/mm)
27 ˚C	2.159	1.669	1.335
75 ˚C	1.668	3.939	3.151
110 ˚C	1.591	6.889	5.511

This finding shows that with increasing temperature, the strength of the polymer decreases, and the deformability increases. Interdomain bonds have less energy than intradomain bonds. This bond causes the ability to deform irreversibly after the impact of mechanical or thermal energy. In general, this process has an entropy characteristic and leads to an increase in the relaxation and regularity of the polymer structure, resulting in the release of thermal energy. Finally, with heating, the bonds between domains of the polymer structure lose elasticity, and more regions between domains lose rigidity. This condition causes the deformability of viscoelastic polymers (Korolev et al. 2021). As the temperature rises, molecular segments move, resulting in free chain movement until the glass transition phase (Grasso et al. 2018). PA tends to endure thermal degradation when exposed to high temperatures, decreasing mechanical properties, including tensile strength.

SEM results (Figure 4) showed that the sample layer at room temperature separated without melting and did not leave any residual material at the edge of the layer after being subjected to tensile load. The absence of temperature in this tensile test has caused the sample layers to separate effortlessly. The sample at a temperature of 75˚C, on the other hand, shows an excess of sample attached to the edge of the layer, indicating that the PA has started to experience thermal degradation or melt after being tested at a temperature of 75˚C. At a temperature of 110˚C, the sample begins to melt and shrink to form an empty cavity on the surface of the sample. The presence of high temperatures when subjected to tensile loads has caused the sample to melt before breaking.



(a)



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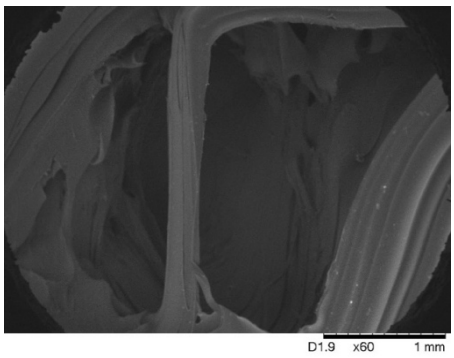


(c)

FIGURE 4. SEM image for tensile fracture sample at temperature (a) 27˚C (b) 75˚C (c) 110˚C

EFFECT OF ELEVATED TEMPERATURE ON DENSITY OF POLYAMIDE

Table 4 exhibits the density result of the tested specimens. Based on the findings, the density of the material decreases after being exposed to increased temperature. According to J.Ling, Moebs & Sanny (2016), materials expand when heated and shrink when cooled. This expansion occurs because the increased temperature causes the atoms or molecules in the material to vibrate more strongly, occupying more space. Interatomic forces hold the elements of solids together. As the temperature increases, atoms absorb energy and vibrate more strongly, causing the bonds between atoms to stretch. This stretching increases the separation between atoms, which results in the expansion of the material. High temperatures cause the material to expand, increasing the volume of the sample while keeping its mass constant. As a result, as the temperature rises, the density decreases.

TABLE 4. The density of polyamide under elevated temperature

Type of test	Density
Flexural test	1.073
Tensile test at 27˚C	1.089
Tensile test at 75˚C	1.075
Tensile test at 110˚C	1.048

THERMAL ANALYSIS (TGA)

Figure 5 depicts the TGA curves of the bending and tensile experiments conducted at three distinct test temperatures. All nylon samples with a single-stage degradation pattern demonstrated thermal stability up to 400˚C. At temperatures between 400˚C and 800˚C, the material begins to degrade rapidly, with a 92% to 98% weight loss in this temperature range attributable to chain dissociation.

According to research, the tendency of PA to absorb moisture contributes to its thermal stability in the early phases of heating up to 400˚C. Consequently, the PA is loosely bound moisture progressively evaporates at this stage, and a mass decrease is observed on the TGA curve (Ray & Cooney 2018). Temperature can result in thermal degradation of polyamides, in which the polymer molecules suffer chemical damage. The fragmentation of the polymer chain has the potential to generate smaller particles. However, exposure to various test temperatures did not substantially alter the TGA curve, indicating that PA did not undergo a significant decrease in molecular weight during exposure to the test temperature. The study discovered that the degradation of nylon led to the production of volatile products that altered the polymer's elemental composition. During degradation, cross-reaction, the formation of volatile charcoal, and aromatic compounds are some of the reactions that occur (Ray & Cooney 2018).

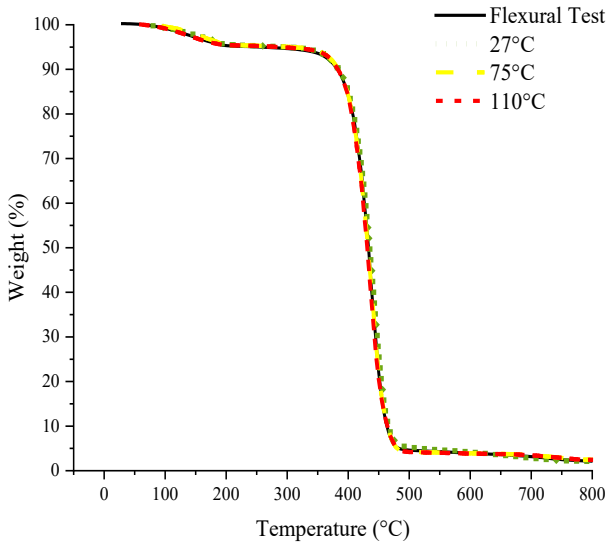


FIGURE 5. Weight loss of Polyamide under elevated temperature

DIFFERENTIAL SCANNING CALORIMETRY (DSC)

The thermogram graph (Figure 6) shows that PA experiences an exothermic reaction or heat release to the environment throughout the heating process up to 800˚C. The baseline increases to a temperature of 450˚C and a baseline shift around 454˚C, indicating that the glass transition occurred at that temperature. An exothermic peak was around 798˚C, indicating an exothermic reaction occurred due to crystallization.

Next, samples at temperatures of 75˚C and 110˚C show the peak of heat flow at a higher degree of gradient compared to temperatures of 27˚C and samples from bending tests. This finding indicates that these two parameters experience a higher crystallization phase transition, resulting in a steeper gradient on the DSC graph. A faster transition rate means faster heat exchange, leading to a more pronounced gradient.

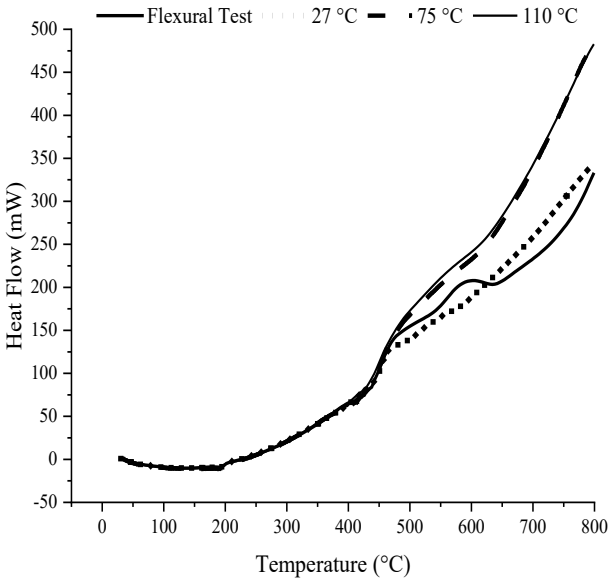
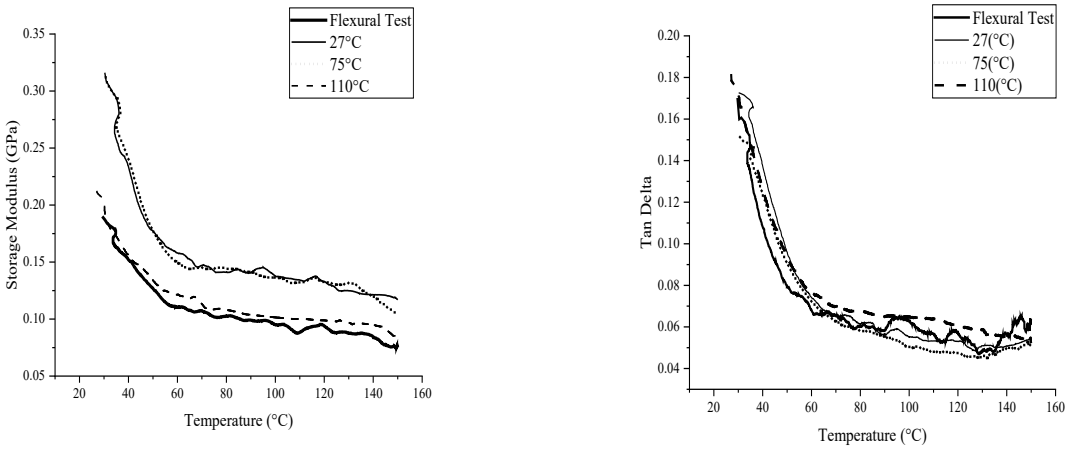


FIGURE 6. Heat flow of polyamide under elevated temperature

DYNAMIC MECHANICAL ANALYSIS (DMA)

The influence of tensile test temperature parameters on the dynamic mechanical properties of FDM printed PA was studied by setting the storage modulus and mechanical damping (ton δ) as response criteria. Figure 7(a) shows a sample from a tensile test at 27˚C has a maximum storage modulus of 0.315 GPa at a temperature of 30.4˚C. On the other hand, the sample from a temperature of 110˚C has the lowest storage modulus, which indicates that this sample has a lower resistance to external force or load after exposure to high temperature. This finding proves that the storage modulus decreases with the increase in tensile test temperature. At high temperatures, polymer chains relax, going from a stressed state to a more relaxed state. This relaxation contributes to energy dissipation and reduces the material's ability to store energy, resulting in a lower storage modulus.

Next, Figure 7(b) shows the effect of increasing tension temperature and DMA temperature on the value of tan δ. The results show that the peak of tan δ for the sample from the flexibility test without temperature is the maximum compared to other samples. For the tensile test sample, the sample at the test temperature of 110˚C shows the maximum tan δ at the beginning of the graph compared to the other samples. A high value of tan δ indicates that the material exhibits significant viscoelastic behaviour. Viscoelastic materials have both elastic (recoverable) and viscous (non-recoverable) properties. The higher the tan δ, the more significant the relative contribution of viscous behaviour to the overall mechanical response of the material. This finding means that a sample at 110˚C dissipates more energy in the heat than stores it elastically.

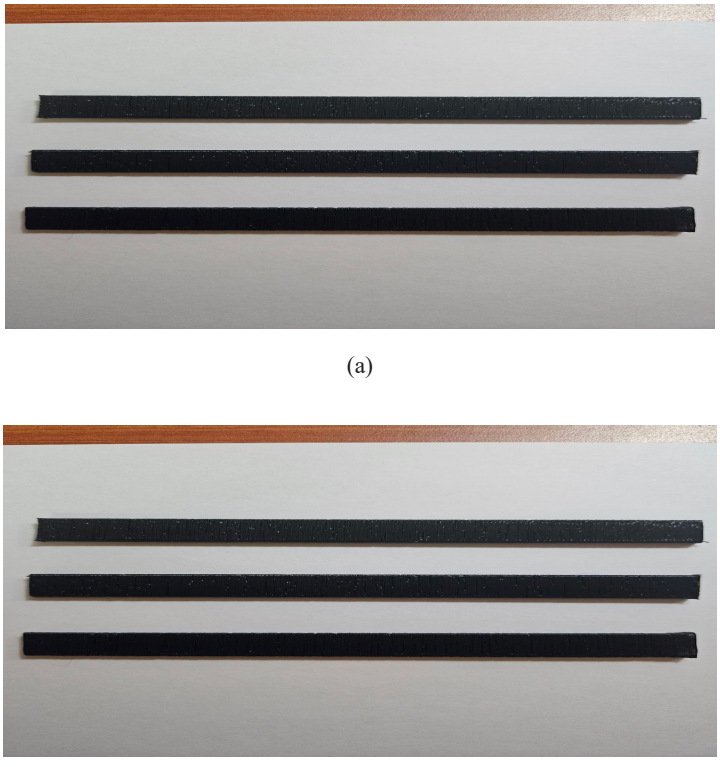


(a) (b)

FIGURE 7. Influence of elevated temperature on DMA (a) Storage modulus (b) tan δ

BENDING ANGLE OF POLYAMIDE

According to the study, the sample begins to change shape quickly after being placed in the oven or with an increase in temperature. After that, the piece changes its shape in the upward convex direction and converges to a constant curvature. The amount of time to achieve this shape change varies for the beam according to the thickness ratio and total thickness (Ali Zolfagharian et al. 2023). Six PA samples with dimensions 127mm×12.7mm×3.2mm were placed in a dry oven at two different temperatures, namely 70˚C and 80˚C to obtain the bending angle. The results (Figure 8) exhibited different results from previous studies when the six samples produced no curves after the 3-hour heating process.



(a)

(b)

FIGURE 8. PA samples that have been heated at a temperature of (a) 70˚C (b) 80˚C

CONCLUSION

Studies related to the influence of layer thickness on bending properties and the effect of increasing temperature on the mechanical properties of polyamide have been successfully conducted. Based on the obtained experimental results and performed analysis, the study found that layer thickness substantially affects the mechanical strength of polyamide, particularly its flexural strength. Increased layer height will elevate bending strength.

Besides, the elevated temperature significantly affects the tensile strength and density of 3D-printed polyamide. With increasing temperature, the strength of the polymer and density diminishes, and its deformability increases.

The results of the TGA test showed that exposure to different test temperatures did not significantly change the TGA curve, indicating that PA did not experience a significant reduction in molecular weight during exposure to the test temperature. The DSC test reveals that higher test temperatures experience a higher crystallization phase transition resulting in a steeper slope on the DSC graph. Besides, DMA analysis exhibits that increasing the test temperature decreases the storage modulus. This finding indicates that this sample has a lower resistance to force or external load after exposure to high temperatures.

The results of this study can be used to predict and understand the effect of layer thickness and temperature on the mechanical and physical strength of polyamide for future advancement in 3D printing technology. However, further research must be conducted mainly related to the influence of layer thickness, raster angle, deformation temperature and recovery temperature on the shape memory effect of 3D-printed polyamide samples.

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DECLARATION OF COMPETING INTEREST

None

REFERENCES

Ali, Z., Hamid, R. J., & Xavier, M. 2023. Multimaterial 4D printing with tunable bending model multimaterial 4D printing with tunable bending model manuscript version: Accepted manuscript. *Smart Materials and Structures*(April) 32(6):065001.

Duc Ngo, T., Kashani, A., Imbalzano, G., Nguyen, K.T.Q. & Hui, D. 2018. Additive manufacturing (3D printing): A review of materials, methods, applications and challenges. *Composites Part B: Engineering* 143: 172–196.

Grasso, M., Azzouz, L., Ruiz-Hincapie, P., Zarrelli, M. & Ren, G. 2018. Effect of temperature on the mechanical properties of 3D-printed PLA tensile specimens. *Rapid Prototyping Journal* 24(8): 1337–1346.

Guessasma, S., Belhabib, S. & Nouri, H. 2021. Effect of printing temperature on microstructure, thermal behavior and tensile properties of 3D printed nylon using fused deposition modeling. *Journal of Applied Polymer Science* 138(14): 1–15.

J.Ling, S., Moebs, W. & Sanny, J. 2016. Thermal Expansion. *University Physics Volume 2*, OpenStax: Houstan, Texas.

Kondo, M.Y., Montagna, L.S., de Melo Morgado, G.F., de Castilho, A.L.G., dos Santos Batista, L.A.P., Botelho, E.C., Costa, M.L., Passador, F.R., Rezende, M.C. & Ribeiro, M.V. 2022. Recent advances in the use of Polyamide-based materials for the automotive industry. *Polimeros* 32(2).

Korolev, A., Mishnev, M., Zherebtsov, D., Vatin, N.I. & Karelina, M. 2021. Polymers under Load and Heating Deformability: Modelling and Predicting. *Polymers* 13(428): 1–14.

Panda, B.N., Shankhwar, K., Garg, A. & Jian, Z. 2016. Performance evaluation of warping characteristic of fused deposition modelling process. *The International Journal of Advanced Manufacturing Technology* 88: 1799-1811.

Ray, S. & Cooney, R.P. 2018. Thermal Degradation of Polymer and Polymer Composites. *Handbook of Environmental Degradation of Materials*, 185–206. Third Edit. Elsevier Inc.: New Zealand.

Samy, A.A., Golbang, A., Harkin-jones, E., Archer, E. & Mcilhagger, A. 2021. Prediction of part distortion in Fused Deposition Modelling (FDM) of semi-crystalline polymers via COMSOL : Effect of printing conditions. *CIRP Journal of Manufacturing Science and Technology* 33: 443–453.

Shahrubudin, N., Lee, T.C. & Ramlan, R. 2019. An Overview on 3D Printing Technology: Technological, Materials, and Applications. *Procedia Manufacturing*, 1286–1296. Elsevier B.V.: Johor, Malaysia.

Tuan Noraihan Azila Tuan Rahim, Abdul Manaf Abdullah & Hazizan Md Akil. 2019. Recent Developments in Fused Deposition Modeling-Based 3D Printing of Polymers and Their Composites. *Polymer Reviews* 0(0): 1–36.

Vishwas, M., Basavaraj, C.K. & Vinyas, M. 2018. Experimental Investigation using Taguchi Method to Optimize Process Parameters of Fused Deposition Modeling for ABS and Nylon Materials. *Materials Today: Proceedings* 5(2): 7106–7114.

Vozniak, A. & Bartczak, Z. 2023. Plastic Deformation of High Density Polyethylene with Extended-Chain Crystal Morphology. *Polymers* 15(66).