Mechanical and Thermal Analyses of Metal-PLA Components Fabricated by Metal Material Extrusion

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Abstract: Metal additive manufacturing (AM) has gained much attentions in recent years due to its advantages including geometric freedom and design complexity, appropriate to a wide range of potential industrial applications. However, conventional metal AM methods have high-cost barriers due to the initial cost of the capital equipment, support and maintenance, etc. This study presents a unique low-cost metal material extrusion (MME) technology. The filaments used have polylactic acid (PLA) as the matrix and metal powders (copper, bronze, stainless steel, high carbon iron, and aluminum) as reinforcements. Using the proposed fabrication technology, test specimens were built by extruding polymer/metal composite filaments, which were then sintered in an open-air furnace to produce solid metallic parts. In this research, the mechanical and thermal properties of the built parts are examined using tensile tests, thermogravimetric-, thermomechanical- and microstructural analysis.

Keywords: Metal Additive Manufacturing, Sintering, Tensile, Mechanical Analysis, Metal Material Extrusion

1. Introduction

Additive manufacturing (AM) is a novel method of manufacturing parts directly from computer models using a layer-by-layer material deposition process. In recent years, AM technology has had an increasing rate of usage in different industries [1]. Due to the wide range of applications, much research is undertaken to investigate the properties of AM parts for practical uses [2]. In general, AM exhibits many advantages including design flexibility, reduced energy consumption and shortened manufacturing times compared with traditional manufacturing methods [3-4].
AM systems are categorized into four main groups of metals [4], polymers [5-6], ceramics [7], and composite systems [8-11]. In the last two decades, metal AM has begun to emerge as an important commercial manufacturing technology [12]. Due to the wide range of applications in automotive, aerospace, oil and gas, and marine, the technology has attracted significant attention [3, 13]. In well-established metal AM technologies such as Selective Laser Sintering (SLS), Selective Laser Melting (SLM), and Direct Metal Deposition (DMD), the energy source fuses or melts the metal powder to produce parts layer-by-layer [14,15], thereby warranting the need for increased electrical energy use, specialist environments (e.g. inert gases) and health and safety considerations for component extraction post-fabrication.

Despite these advantages, the more established metal AM methods are expensive and require sophisticated machines and facilities for subsequent post-processing [16]. Therefore, new technologies may help to reduce the final cost of creating metal AM parts. MME is a newly proposed metal AM technology based on the very popular Fused Deposition Modeling (FDM) process and with lower cost compared to the well-established, existing metal processes [17]. The filament is composed of polymer and metal powder. The part is extruded and turns into pure metal by undergoing secondary debonding and sintering processes. After the debonding and sintering process, a part with almost 100% solid metal is obtained [18]. The low-cost metal process described in this work can be conducted using conventional low-cost desktop systems to produce parts [19].

Research is being conducted to investigate the properties of parts produced with low-cost metal AM processes. Hwang et al. [20] and Masood et al. [21] showed that as the metal fillers in metal-polymer composite materials increase, the tensile strength of the composite material is reduced and the thermal conductivity of the metal-polymer composite increases significantly. Terry et al. [22,23] studied the dimensional changes and microstructure analysis of sintered and unsintered parts made with low-cost metal AM technology. Some researchers used the FDM process to manufacture parts, which were then debonded and sintered in a protective gas environment to form complete metal parts. However, due to the porous structure, it is observed that the tensile strength is not as good as, for example, SLM parts [24].

This research introduces low-cost AM technology for the fabrication of metal parts with low price, easy manufacturing process, and with mechanical properties sufficient for a wide range of engineering applications. Also, the effects of printing parameters on mechanical and thermal properties were investigated. The filaments are composed of polymer (PLA) and metal powders from Copper (Cu), Bronze (Br), Stainless Steel (SS), High Carbon Iron (HC), and Aluminum (Al). For this study, mechanical characterization focused on Cu-PLA composite material whereas thermal characterization focused on Cu-, Br-, SS-, HC-, and Al-PLA composite materials. Figure 1 shows the different steps of operation for this low-cost metal AM process including modeling, file conversion, component slicing, filament warming, extrusion, sintering and post-processing.
2. Materials and Methods

Filaments used in this study were purchased from the Virtual Foundry Company [25]. The filaments were composed of PLA and metal powders as described previously. Bonding agents were used in the filaments to improve the strength between the polymer and metal phases. Weight percentage (%) of Cu, Br, Al, HC, and SS in filaments was 90, 88, 65, 75 and 85, respectively. Those containing Cu, Br and SS were 3 mm in diameter while those containing HC and Al were 1.75 mm in diameter.

Ultimaker S5 (UMS5) and Raise 3D pro2 (R3D) 3D printers were used to build test specimens. UMS5 is a Bowden-drive printer that utilizes 2.85 mm diameter filament. R3D, on the other hand, is a direct-drive printer that utilizes 1.75 mm diameter filament. The test specimens were printed with three different layer heights of 0.1, 0.2 and 0.3 mm and a printing speed of 10 mm/sec. The printing temperature was increased to 230 °C to reduce the viscosity of PLA and increase the flow rate from the 0.4 mm extrusion nozzles. Table 1 shows the printing parameters used in this study.

| Parameter               | Value           |
|-------------------------|-----------------|
| Base material           | PLA             |
| Supports                | No supports     |
| Skirt line count        | 3               |
| Infill density          | 100 %           |
| Wall layers             | Maximum         |
| Printing temperature    | 230 °C          |
| Build plate temperature | 60 °C           |
| Printing speed          | 10 mm/s         |

Instron 5582 Universal Testing Machine was used to measure the tensile properties i.e., ultimate tensile strength (UTS) and elastic modulus (E). The tests were conducted in accordance with the American Standard Test Model (ASTM) D638 requirements [26]. Microstructural morphology and
build quality of the specimens were examined using an optical microscope model Nikon SMZ1500 (Tokyo, Japan). Thermogravimetric analysis (TGA) was conducted using a TA Instruments Q600 (Delaware, USA) to measure the thermal decomposition temperature, thermal decomposition rate, true sintering temperature, and true weight percentage of the metal powders. Thermomechanical analysis (TMA) measured material deformation under the controlled thermal conditions. The test was conducted using TA instruments’ TMA Q400 (Delaware, USA) to study the coefficient of thermal expansion (CTE) of parts built by MME. Test specimens were cube-shaped with a side length of 7 mm, fabricated with 0.1 mm layer height and contour infill pattern.

3. Results and Discussion

3.1. Tensile Test

The tensile properties of Metal-PLA (MPLA) specimens were studied. The effects of extrusion parameters including layer thickness, and sintering temperature on tensile strength, elastic modulus and yield strength were also investigated. Test specimens examined were manufactured in accordance with ASTM D638 requirements.

Tensile test specimens from Cu-PLA filaments were built with layer thicknesses of 0.1, 0.2 and 0.3 mm and sintered in Lindberg Blue M furnace (Massachusetts, USA) at 1065 °C to remove bonding agents and accelerate fusion between metal powder particles. The sintered specimens were manufactured as per ASTM E8/E8M guidelines for tensile testing of powder metallurgy specimens. In terms of layer thickness, 0.1 mm is the minimum that can be extruded without nozzle abrasion and clogging, 0.2 mm is the machine’s default, whereas 0.3 mm is commonly used based on nozzle size.

Table 2 compares data from PLA, Cu-PLA, and sintered Cu-PLA built in this study at 0.1 mm layer height with annealed Cu [27]. Although the tensile properties of PLA are higher than Cu-PLA, it is evident that sintering improved the latter immensely, with values comparable to annealed Cu [27]. Data shown in Table 3 for sintered Cu-PLA imply a decrease in UTS and E values as layer thickness increases. The stress-strain curves of the specimens are shown in Figure 2. The decline in mechanical properties in relation to increasing layer thickness could be attributed to the density of metal particles in the bead. Figure 3 shows a schematic of the effect of layer height on metal particles in the bead: as layer thickness reduces, the density of metals in the bead increases.

| Material                  | UTS (MPa) | E (GPa) | Yield Stress (MPa) |
|---------------------------|-----------|---------|--------------------|
| PLA                       | 66        | 3.5     | 48                 |
| Cu-PLA                    | 22        | 1.4     | 18                 |
| Cu-PLA sintered at 1065C  | 188       | 101     | 55                 |
| Annealed Cu [22]          | 210       | 110     | 33.3               |

Table 3. Tensile properties of sintered Cu-PLA specimens

| Layer Height (mm) | UTS (MPa) | E (GPa) | Yield Stress (MPa) |
|-------------------|-----------|---------|--------------------|
| 0.1               | 188       | 101     | 55                 |
| 0.2               | 166       | 39      | 36                 |
| 0.3               | 153       | 15      | 39                 |
Figure 2. Stress-strain curves of sintered Cu-PLA built with different layer thickness.

Figure 3. The effect of layer height variation on the metallic concentrations in deposited beads.

Test results show that before sintering, the MPLA specimens have poor mechanical properties which limit any potential applications in this form. However, after being subjected to the sintering process, the mechanical properties of the MME Cu specimens are greatly improved. The mechanical properties of MME components are however lower than the more traditionally produced Cu parts, but still have higher mechanical properties compared with SLM, and Jet Binder produced parts. Overall, it can be concluded that the MME specimens have mechanical properties within an acceptable tolerance/range of the standard engineering materials.

3.1. Thermomechanical Analysis (TMA)

Table 4 shows the CTE values of different specimens at distinct x, y, and z printing directions. As shown, metal inclusion reduces the CTE value of PLA, for which Cu-PLA recorded the lowest value as compared to other specimens. Also, the CTE value for MPLAs are noted to be higher than traditional metals [27]. The purpose of furnace sintering is to employ the thermal expansion of the metal powder to push out the air in the structure to form a solid part. The analysis described herein was conducted in two different directions of the specimens: x/y - and z - axis. The CTE values
obtained for MPLA specimens in the $z$ direction are higher than those in the $x/y$ direction. This is possibly due to the interface between layers. Because layers are perpendicular to the $z$ direction, an increase in interface can cause expansion in the $z$ direction more than the $x/y$ direction.

### Table 4. CTE of different filaments in the different extrusion directions

| Material       | Average CTE mm/ (m C) |
|----------------|-----------------------|
|                | Direction             |
|                | $x/y$ direction | $z$ direction |
| PLA            | 0.07906               | 0.07983       |
| Cu-PLA         | 0.05646               | 0.06311       |
| Al-PLA         | 0.06927               | 0.07244       |
| HC-PLA         | 0.06413               | 0.06980       |

#### 3.3 Thermogravimetric Analysis (TGA)

TGA is a thermal analysis method in which the mass of a sample is measured over time as temperature increases at a defined rate. The tests for Cu-PLA, Br-PLA, SS-PLA, Al-PLA, and HC-PLA specimens were conducted to comply with ASTM E1131 [28] in the temperature range from room temperature to 500 °C and a constant heating rate of 10 °C/min.

Each test was repeated (2x) for reliability. Figure 4 shows the TGA curves of the five MPLA filaments examined. Cu-PLA recorded the highest thermal resistance followed by Br-PLA and SS-PLA. The lowest thermal resistance was recorded for Al-PLA. In this study, TGA provided detailed information on the degradation temperature and rate of decomposition of the materials within the temperature range of 250 - 400 °C at 2 h. Each curve reveals the mass loss per degradation temperature from which the degradation rate was calculated: Cu-PLA and Al-PLA composite materials produced the lowest and highest degradation rate, respectively. MPLA decomposition temperature and residues are listed in Table 5. The decomposition temperatures are in the range of 230 to 400 °C. All the curves plateaued out after ≈ 430 °C until the end of the test. The horizontal part of the curve at high temperature confirms the residue (metal weight percentage) content. The data presented in Table 5 also corroborate the assertion that the thermal degradation temperature of PLA is higher than MPLAs [29].

![Figure 4. TGA plot of MPLA specimens](image-url)
Table 5. Thermal degradation temperature of MPLAs and PLA

| Material | Thermal Degradation Starting Temperature (°C) | Full Degradation Temperature (°C) | Weight % |
|----------|-----------------------------------------------|-----------------------------------|----------|
| Cu-PLA   | 300.87                                       | 437.45                            | 87.28    |
|          | 293.73                                       | 399.49                            | 85.51    |
| Br-PLA   | 311.21                                       | 405.59                            | 85.98    |
|          | 242.52                                       | 439.45                            | 83.60    |
| Al-PLA   | 279.41                                       | 420.54                            | 60.49    |
|          | 271.37                                       | 420.65                            | 60.45    |
| SS-PLA   | 252.08                                       | 424.64                            | 83.44    |
|          | 227.33                                       | 408.27                            | 83.80    |
| HC-PLA   | 267.83                                       | 422.10                            | 75.96    |
|          | 262.95                                       | 420.65                            | 76.36    |
| PLA      | 350                                           | 400                               | 0.1      |

3.4 Microstructural Analysis

The cross-sectional view of unsintered Cu-PLA specimens is shown in Figure 5. Two different phases for metal and polymer are visible with the bulk of the space filled by PLA. In the micrograph, the dark Cu particles (10 to 50 µm) are interspersed in the matrix.

![Cross-sectional view of Cu-PLA specimens](image)

Figure 5. Cross-sectional views of Cu-PLA specimens

4. Conclusions

MME demonstrates a relatively low cost (in the context of equipment, materials, and maintenance) manufacturing option as compared to established metal AM processes. This research study thus presents a novel method that can be explored to fabricate affordable metallic parts. In this study, test specimens were prepared from composite materials using PLA (matrix) and Cu, Al, Br, HC, and SS (reinforcements) formulations. The furnace sintering process induced material shrinkage which ensured that the final parts exhibited improved mechanical properties. In TMA, the printing directions influenced the CTE of MPLAs. TGA confirmed that thermal degradation of the base polymer commenced at ≈ 250 °C.

Due to the COVID-19 pandemic situation, the mechanical tests for sintered specimens were not fully completed. Future work will aim to improve the physico-mechanical properties of the parts (e.g.,...
reducing the voids created during sintering) and assess their suitability for different applications using for instance, fatigue and dynamic mechanical analysis to further characterize them. While the basic mechanical/thermal properties and sintering shrinkage ratios are gradually improved, a statistical model can be created for predicting dimensional changes of parts and their mechanical properties at different sintering temperatures.

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