Modeling and Microstructure Analysis of Three-Dimension Direct Writing for Zn-Al Alloy

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Abstract. This paper presents a systematic numerical experimental investigation of single-track forming of 3D metal melting direct writing. A forming track width was fitted by multivariate nonlinear regression method, and the numerical model had a fair level of uniformity with the experiment results. Two phase flow model based on volume-of-fluid (VOF) method was adapted to build up the coupled calculative models for direct writing deposition and solidification. The results show that when the forming track width approximately equals to the solidification layer width, it is the optimum time for forming the next layer. Based on the above research, the pre-remelting time was proposed, which is helpful to create next layer forming quality, and could also be used to guide the creation of a proper default of process parameters.

Keywords: Metal melting direct writing, Metal additive manufacturing, 3D printing, Dimension modeling, Microstructure analysis

1. Introduction
Additive manufacturing (AM) of metal is a numerical forming manufacturing technology based on the dispersion-deposition principle. Specifically, after processing the raw materials by laser, electron beam, electric arc or compound heat source melting processes, the metal components are stacked by the melted metal via CAD modeling. Comparing with traditional metal parts forming, metal AM is advantageous in various aspects: no die needed, short lead time, and high flexibility and customizability on small scale manufacturing of metal dies and components. Recently, some mature metal AM technologies including selective laser sintering (SLS), electron beam melting (EBM), wire arc additive manufacturing (WAAM) and metal micro-droplet deposition manufacturing (MDDM), have been introduced in aerospace, biomedical and other fields\cite{1-3}.

3D metal melting direct writing (3D-MMDW) is a branch of metal AM system in the industrial manufacturing area which was firstly proposed by China Academy of Machinery Science and Technology\cite{4}. Induction heating, resistance heating or other heating technologies are applied in 3D-MMDW for metal melting process. As shown in figure 1, the melted metal will be pressed out from nozzle and performed the following processes layer by layer with designed route: jetting, depositing,
solidifying and compounding, the designed 3D metal component is formed incrementally from bottom to top. Different from laser or electron beam AM, a high energy heat source is not necessary to conduct the writing, this could be positive to industrial manufacturing. Additionally, 3D-MMDW can form multi-material gradationally, reduce more cost and enhance more forming efficiency that binder jetting (BJ) technology is short on.

3D-MMDW integrates computer numerical control (CNC), casting, welding and other technologies, the reaction alters inner energy and outer shape by depositing and solidifying melted materials on the substrate. While processing, the microstructure and size of the designed component are impacted, it will impinge the properties and precision further. Therefore, the key point (also the difficulty) of the direct writing technology is to carry on a series of researches in molten metal forming process and figure out the relationship between deposition and solidification, to precisely control the size and properties of designed components.

Researchers have devoted to research on molten metal dropping, extending and solidifying of additive manufacturing process. Pasandideh [5] and Mostaghimi established molten droplets model based on conservation of momentum and linear theory that coupled with experiment result ideally. Zeng and his team who are from Northwestern Polytechnical University used computational fluid dynamics (CFD) software Fluent to simulate the interaction and solidification of a single molten droplet in the course of depositing process, and worked out the regularity that how process parameters will influence the molten droplet formation and solidification under precise control. Li and Liu [7] applied the VOF method to simulate the temperature variation of molten droplets while dropping colliding and spreading, and collected temperature gradient data. Song and his group combined simulation with BJ experiments, and made corresponding strategies to control jetting variables, thus improve work pieces quality.

It is worth to mention that the above researches about molten metal deposition fluid-solid coupling process are all setting dispersed droplet as the minimum unit, in this case, it cannot match to the deposition-solidification process of melted metal of melting direct writing appropriately. Besides, the simulations were mostly relative to two-dimension computing which lacked the consideration of horizontal deposition. The exclusive reports and researches of molten metal’s formation and solidification on 3D-MMDW have never been worked out. In Yang’s thesis [9], researchers did pre-research on 3D-MMDW, and they found the relations among Sn-Bi alloy process parameters, single track forming size and 3D forming precision, reasonable process parameters range was predicted based on the findings. However, they mainly focused on low melting point alloys direct writing technologies and devices, a profound study of direct writing processes is still required to be made.

This report is aiming at Zn-Al alloy single track melting forming direct writing, under the restriction of different process parameters, size model of single-track forming will be set up. The finite element method (FEM) is introduced to simulate single track forming fluid-solid coupling process in order to figure out principle of size alteration and temperature distribution. In the end, angling towards
microstructure formation, further analysis and verification of molten metal temperature alteration in direct writing is conducted which lays foundations for shape and properties precise control.

2. Materials and Experimental Procedure
Self-developed 3D-MMDW equipment and Zn-Al alloy are the preparations for this single-track forming experiment. Table 1 and 2 illustrate the compositions and thermal properties of Zn-Al alloy.

| Table 1. Zn-Al Alloy Compositions (wt%) |
|---------------------------------------|
| Alloy Designation | Al | Cu | Mg | Zn |
| ZnAl4Cu1Mg | 3.5–4.5 | 0.75–1.25 | 0.03–0.08 | Remaining Composition |

| Table 2. Zn-Al Alloy Thermal Properties |
|----------------------------------------|
| Thermal Properties | Value |
| Density (kg/m³) | 5000 |
| Specific Heat Capacity (J/kg·K) | 525 |
| Thermal Conductivity (W/m·K) | 125.5 |
| Viscosity (Pa·s) | 0.004 |
| Latent Heat of Fusion (kJ/kg) | 128 |
| Solidus (K) | 649 |
| Liquidus (K) | 757 |

Zn-Al alloy is heated and melted in the crucible, sprayed out from metal nozzle under the pressure of protective gas, and then deposited on the motional substrate. The initial temperature of deposition is 773K, and the initial temperature of substrate is 303K. The single-track forming experiment was performed by following the designed moving path under different nozzle diameters (D) and nozzle-substrate distances (H). Solidified liquid column as shown in figure 2 was set as basic unit for further test and analysis. Vernier caliper was used to measure the width of the forming track, the mean of 12 measurements was selected as the standard size.

Based on the experimental size data, the free surface of molten metal could be traced via the VOF method in Star-CCM+ fluid dynamics simulation software. Similarly, the solid-liquid transition of melt could be traced by the melting-solidification model as well.

Zeiss scanning electron microscope (SEM) was introduced to observe the microstructure and phase composition of forming unit, and then the deposition-solidification process of melt was analyzed.

Figure 2. Solidified Liquid Column Samples of Single-track Forming
3. Result and Discussion

3.1. Size modelling analysis of formation

In the direct writing technology, the sample of single-track forming was defined as the basic forming unit. During the direct writing process, the sample would influence forming precision of the 3D entity accumulatively. The connection among nozzle diameter (“D”), the height between nozzle and substrate (“H”), and the width of the single track (“w”) (Figure 3) were demonstrated by a mathematical model base on multiple non-linear regression method.

Set “w” as response variable, “D” and “H” as predictor variables, the regression equation was formulated:

\[ w = 2.63D + 0.31H + 3.78D^2 - 0.004H^2 + 0.25DH - 4.28 \]

Figure 3. The Cross Section Size of Sample

The regression function image is plotted in Figure 4. For testing the practical value and to what extend the regression equation accord with the real situation, carrying out a significance test of regression is essential. In F-test, the value of F is 45.15 after calculating, which is much greater than 4.69 (when \( \alpha = 0.01, F_{(5,14)} = 4.69 \)). This phenomenon illuminates that “D” and “H” owns significant regression relation with “w”. Variance test value R²=0.9207 shows the high fitting degree of the equation. Table 3 shows the fitting value and experiment results, proves that the resulting regression equation is a credible model of the data.

Figure 4. The Regression Function Image of Forming Characteristic Parameters
Table 3. Comparison of Fitting Value and Experiment Results

| Experiment | Predictor Variables | Response Variable | Fitting Width w(mm) | \( \delta \) (mm) |
|------------|---------------------|------------------|---------------------|------------------|
|            | Diameter D(mm) | Height H(mm) | Width w(mm) | |
| 1         | 1.0   | 30            | 13.71333        | 15.33            | 1.616667       |
| 2         | 1.0   | 40            | 18.97667        | 18.13            | -0.84667       |
| 3         | 1.0   | 50            | 20.22           | 20.13            | -0.09          |
| 4         | 1.0   | 60            | 23.17667        | 21.33            | -1.84667       |
| 5         | 1.0   | 70            | 20.93           | 21.73            | 0.8            |
| 6         | 0.8   | 30            | 11.91333        | 11.9432          | 0.029867       |
| 7         | 0.8   | 40            | 13.33333        | 14.2432          | 0.909867       |
| 8         | 0.8   | 50            | 14.41333        | 15.7432          | 1.329867       |
| 9         | 0.8   | 60            | 18.01           | 16.4432          | -1.5668        |
| 10        | 0.8   | 70            | 13.76333        | 16.3432          | 2.579867       |
| 11        | 0.6   | 30            | 11.25           | 8.8588           | -2.3912        |
| 12        | 0.6   | 40            | 9.836667        | 10.6588          | 0.822133       |
| 13        | 0.6   | 50            | 12.21           | 11.6588          | -0.5512        |
| 14        | 0.6   | 60            | 11.19667        | 11.8588          | 0.662133       |
| 15        | 0.6   | 70            | 11.72333        | 11.2588          | -0.46453       |
| 16        | 0.4   | 30            | 5.913333        | 6.0768           | 0.163467       |
| 17        | 0.4   | 40            | 6.323333        | 7.3768           | 1.053467       |
| 18        | 0.4   | 50            | 6.743333        | 7.8768           | 1.133467       |
| 19        | 0.4   | 60            | 7.356667        | 7.5768           | 0.220133       |
| 20        | 0.4   | 70            | 7.316667        | 6.4768           | -0.83987       |

In terms of the process parameters, on an adequate confidence level, it was not difficult to predict the wall thickness of the designed component. On the contrary, the wall thickness owned the referential significance for the parameters. In this case, both machining allowance and produce time would be reduced.

3.2. Numerical simulation of forming process

As shown in Figure 5, the geometrical model is set up and meshed in Star-ccm software. The initial molten metal region is single-tracked, the rest of the region is defined as the initial air region, and the both of them are set as Eulerian two-phases region.

VOF method is introduced to trace the free surface of metal deformation, and the metal-air interface can be located via VOF governing equation by solving the volume-fraction value \( F \) of the metal in the meshing elements. VOF governing equation is as follows:

\[
\frac{\partial F}{\partial t} + (V \cdot \nabla)F = 0
\]

 Defined the metal and air region as \( \Omega_m \) and \( \Omega_a \) respectively, defined the free surface as \( \Gamma \), and the element coordinate as \( (i, j) \), then the new free surface determination can be figured out by \( F \) value, thus the metal shape deformation can be traced.

\[
F = \begin{cases} 
1, & (i,j) \in \Omega_m \\ 
0 < F < 1, & (i,j) \in \Gamma \\ 
0, & (i,j) \in \Omega_a 
\end{cases}
\]
Meanwhile, in order to work out the free surface of solidified metal and trace the metal phase transition, the solidification-melting model is applied in this analysis.

![Regions of Multiphase Flow model](image)

**Figure 5.** Regions of Multiphase Flow model

Set the initial temperature of the liquid region as 773K, the temperature of the substrate and the air is 303K. The shape deformation and the solidifying process of the single-track molten metal during a period are demonstrated in figure 7. Zn-Al volume fraction distribution on the cross section are showed at the left of figure 7, and the solidified volume fraction distribution on cross section is at the right.

The contact area of the metal and substrate are solidified swiftly, and an extremely thin solidified layer is formed on the interface of metal and air. Due to the gravity, the melt overcomes the impedance of surface tension, gradually deforms down and shapes into an ellipse from circle.

The surface of solidified metal contacting with the substrate, sprawls out and moves up. While the surface of solidified metal contacting with the air moves in.

The width of the solidified metal is \( w_s \), the value is growing up over time, the spraying width of the melt increases as well. As illustrated in Figure 7 (a) (b), after 1.45 seconds, the value of \( w_s \) is equal to the value of \( w \), displayed in Figure 7 (c). The deformation tends to stability at that point. The values of the widths in (c) (d) (e) are almost the same. The solidifying process is still in progress until Zn-Al has fully solidified.

3D-MMDW technology is the process of manufacturing components layer by layer. If the solid fraction of the first layer is not high enough, when the following layer covers on the former layer, it will lead to collapse and excessive deformation. On the contrary, if the first layer solidified proportion is pretty high, due to large temperature difference, the next layer cannot attach to the last layer, which will cause lack of layers combination. When the first layer is in semi-solid state, as shown in figure 7, the contour of metal is kept ideally with fair liquefied proportion, it is the optimal timing for layers combination. \( t_r \) is defined as remelting preparatory time, when \( w \) value is equal to \( w_s \) value as solidification procedure processes, it is the optimum to maintain the forming process of next layer. The \( t_r \) value cannot be collected with experiments directly, calculation is essential. In terms of D and H, \( w \) can be figured out, \( t_r \) will be got from simulation calculation, and then the forming parameters of 3D-MMDW have been resolved for further qualified printing.
3.3. Temperature distribution and Microstructure analysis

The sample of the single-track forming could be divided vertically into three areas, named “A”, “B” and “C” area from top to bottom (Figure 7). During the melt extension process, the heat exchange among melt, substrate and air increased the environment temperature. Because the thermal conductivity of Zn-Al alloy and substrate is higher than that of metal and air, hence from temperature field distribution pattern, the melt temperature gradient of upper area was less than that at alloy-substrate contact area which would lead to the existence of various solidification microstructure in different areas. In figure 7, “A” area was the direct contact area which illustrates increasing temperature gradient from substrate to inner melt. “C” area was the largest melt section with high thermal capacity which was similar to the hot spots of casting, thus the temperature was relatively high and distributes evenly. “B” area was located at the arc of the melt with large air contact area, the temperature gradient had a decreasing trend from outer melt to inner melt.

Zinc alloy sample was made by the direct writing technology, and as-cast microstructure was formed. The microstructure was composed of primary phase $\alpha$ and eutectoid phase $\alpha + \eta$. The pale branch shape $\alpha$ phase is solid solution formed by Zn dissolving in Al, similarly, the gray $\eta$ phase is solid solution that formed by Al dissolving in Zn. “A” area has the most obvious temperature gradient and the fastest cooling rate, presenting directional fine dendrites. Although the grain size is small, the apparent orientation of the grains will affect the mechanics performance of the specimen in different directions. “C” area is located inside the molten metal, with larger grain size and relatively uniform equiaxed
crystals, which is similar to the as-cast structure obtained by sand casting of Zn-Al alloy. This zone is located in the hot-spot section of single-track forming, the average mechanical and physical properties of the part are mainly determined by the microstructure of the zone; the “B” area is located at the outer layer of the single-track forming metal. The grain size is medium, and presents a dendritic state. As the acceleration of cooling rate, the crystal grain size decreased, the microstructure got refined. If the speed of solidifying was over the limit, the equilibrium state would be broken which would lead to alloy composition segregation. The microstructure could be improved by adjusting the solidification rate.

Figure 7. Temperature Distribution and Microstructure at Different Areas (200x)

4. Conclusion
(1) Width in single track forming is a key parameter that is highly related to the thin-walled parts’ homogeneity and precision of melt direct writing forming. The forming track width was fitted by Multivariate nonlinear regression method, and the fitting results had a fair level of uniformity with the experiment results. The results showed that the nozzle diameter (“D”) was the most influential parameter to the forming track width, followed by the height between nozzle and substrate (“H”).

(2) VOF was applied to simulate the deformation process of metal melting direct writing. Comparing with the experiment results, different cooling levels brought different temperature gradients, hence three different solidification microstructures were generated vertically in the liquid column.

(3) Pre-remelting time was proposed in the direct writing technology. When the forming track width approximately equals to the solidification layer width, it is the optimum time for forming the next layer. The pre-remelting time can be used to guide the creation of a proper default of initial temperature, heat transfer boundary condition, substrate motion and other process parameters.

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