Residual stress mitigation in directed energy deposition

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**A R T I C L E   I N F O**

**Keywords:**
Directed energy deposition  
Residual stress  
Immiscible alloys  
Additive manufacturing

**A B S T R A C T**

Directed Energy Deposition (DED), a class of additive manufacturing techniques, has seen rapid growth over the last decade for potential applications in aerospace, medical devices, and energy systems. Despite notable progress in the research and development of AM, control and mitigation of residual stress during DED remains a challenge. In this work, we propose a novel approach that can be used for the mitigation of residual stresses in additively manufactured components. Specifically, we propose to mitigate the residual stress state of as-deposited components using alloy design, engineering of solid-state transformations, and the introduction of both hard and soft metallic phases. We demonstrate this strategy with a model system consisting of pure Fe and Fe–Cu. Experimental results indicate that residual stresses can be successfully manipulated by adjusting the alloy composition as a soft metallic phase can accommodate plastic deformation. Moreover, our findings suggest that the solid-state transformations experienced by the Fe and Fe-rich phases contribute to the observed differences in magnitude and location of residual stresses. This study is the first to suggest using residual stress as an engineering criterion in the design of alloys for metal additive manufacturing.

1. Introduction

Metal additive manufacturing (AM), often referred to as metal 3D printing, is a family of disruptive manufacturing technologies that allow for rapid, on-demand fabrication of high-quality near-net-shape metal components. Metal AM has been successfully adopted by the medical [1, 2] and aerospace [3,4] industries, where custom, geometrically complex metal parts are required. The ever-increasing number of patents and journal publications in metal AM show how interest in this topic, from both industry and academe, continues to increase. Such a keen interest from the metallurgy and manufacturing communities can be attributed to metal AM’s capability to significantly reduce lead times, enabling efficient and rapid iterations in design and testing [5]. Additionally, metal AM allows for considerable part count reductions in assemblies [6–8], as well as light-weighting through part geometry optimization [9]. In contrast to conventional manufacturing processes, in metal AM, a part is built up from the feedstock material (e.g., powder or wire) layer-by-layer based on a computer aided design (CAD) model. Inspection of the literature shows powder bed fusion (PBF) and directed energy deposition (DED) are perhaps the most widely used techniques. In a typical PBF process, a thin layer of feedstock powder is spread uniformly across the build platform by a roller or a re-coater, and a focused energy source is scanned along the CAD-defined path, melting the powder particles. The build platform is then lowered, and the process is repeated for the next layer. In DED, the feedstock material is delivered directly to the melt pool formed by a focused energy source (laser/electron beam or arc plasma). While PBF commonly has higher dimensional accuracy and yields parts with smaller surface roughness, in comparing DED and PBF the following points are frequently made: 1) DED can be used for higher deposition rates (up to 2.5 kg/h for DED versus 0.25 kg/h for PBF [10]), 2) DED can be applied for multi-material deposition (e.g., *in situ* alloying [11]) as well as the fabrication of functionally graded structures [12,13], 3) DED can be used for the preparation of coatings [14,15] and repair of damaged components [16,17], and finally, 4) DED can be applied for the fabrication of large (>1000 mm³) part volumes [18].

DED has been compared to welding; however, it also presents a variety of unique technical and scientific challenges. Microstructures of DED-deposited alloys usually exhibit highly elongated grains and preferred crystallographic orientations [19], leading to significant non-uniformities in mechanical properties [20]. Moreover, the relatively high cooling rates of up to 10⁶ K/s [21–24], in combination with the inherent layer-by-layer nature of DED exposes the material to remelting and thermal cycling which can lead to phase transformations [25–27,28,23,29–32] as well as the formation of detrimental residual...
stresses and changes to the microstructure. As a consequence, the residual stresses that are introduced by DED can be highly nonuniform and often have high gradients up to ~10^7 MPa/mm. Residual stresses in DED-fabricated parts, which often have tensile character, can cause a multitude of issues, including: distortion, cracking, delamination of parts from the substrate, and early crack propagation under cyclic loading, thereby leading to early failure. Understanding, predicting, and mitigating residual stresses in DED parts is, therefore, of great importance.

A review of the published literature demonstrates that heating of the substrate can be effectively used to mitigate residual stress. Applying heat during deposition can minimize residual stresses during printing. In related studies, Corbin et al. showed that preheating the substrate to ~400 °C reduces the substrate distortion that occurs during deposition of the first layer by 27%. Similarly, Lu et al. formulated a 3D thermo-mechanical finite element (FE) model and reported that when both substrate preheating and chamber heating are implemented, residual stresses and distortions decrease by 80% in 90%, respectively. Finally, Vasinont et al. used a 3D thermo-mechanical model for LENS® and found that baseplate preheating reduces the residual stress by a significant amount, as high as ~40%. In summary, these studies demonstrate that substrate, build chamber, and component pre-heating can be used for residual stress mitigation, although complete residual stresses elimination is generally not accomplished, and further post-processing is sometimes required.

Optimization of the scan strategy can also be used to mitigate residual stress. In related work, Denlinger et al. studied the effect of interlayer dwell time on part distortion in Ti-6Al-4V and Inconel 625 L-DED parts. Their results confirmed that increasing interlayer dwell time from 0 to 40 s increases cooling rate during deposition and reduces residual stress from ~710 MPa to ~566 MPa in Inconel 625. However, they also found that increasing dwell time from 0 to 40 s during printing of Ti-6Al-4V led to an increase in residual stress from ~98 MPa to ~218 MPa. These results suggest that the formation of residual stresses is material-dependent, reflecting the complex interplay between residual stress and microstructure. In particular, the differences in behavior of Inconel 625 and Ti-6Al-4V depend on the phase transformations that occur during AM. In related studies, Woo et al. studied the influence of scan strategy on the residual stress in functionally-graded materials (FGM) using L-DED. Their results show that stress can be reduced from ~950 MPa for 0° hatch rotation to ~680 MPa for 90° hatch rotation, and then further reduced to ~430 MPa with the island or “checkerboard” approach. Similarly, in related work, Yu et al. used a fractal scanning strategy, where the layers were scanned following a Hilbert curve. Their results showed that components printed with this strategy exhibited less substrate deformation than those prepared using conventional scan strategies. Ma and Bin confirmed that fractal scan strategies lead to ~55.7% lower distortions than those produced by a serpentine scan strategy in selective laser sintering. Interestingly, Strantza et al. found that employing a widely popular “island” or “checkerboard” scan strategy in LPBF of Ti-6Al-4V in place of continuous scanning throughout the part can increase residual stresses, especially in surface areas of the printed component. They also observed higher cut-off deflections in the part printed using the “island” scan strategy. Overall, these studies demonstrate that scan strategy can have a significant influence on the residual stresses and distortions in DED.

Thermal treatment of the fabricated component can also decrease residual stresses. First, the formed workpiece residual stresses after heat treatment of Inconel 625 parts in combination with compression testing with in situ neutron diffraction was reported by Wang et al. Their results quantified the extent of stress relaxation and reported macroscopic stress as well as local stress in grains with different orientations. The results indicate that at the same temperature and applied strain, additively manufactured Inconel 625 had a higher stress relaxation rate and lower peak and plateau stresses than conventionally processed Inconel 625, due to different texture and grain sizes in these two materials. Similarly, Wang et al. employed neutron diffraction and demonstrated that residual stress in laser-based DED-printed Inconel 625 parts can be mitigated via heat treatment at 870 °C for 1 h. These studies suggest that post-print heat treatment can be used as an effective approach for residual stress relief, but that specific alloy compositions may yield different results, depending on phase transformation, for example.

As discussed above, the most popular stress mitigation techniques in metal AM are based on adjusting process parameters, such as substrate/chamber temperature, scan strategy, or interlayer dwell time. These approaches are not capable of entirely eliminating residual stresses or converting detrimental tensile stresses into beneficial compressive stresses. An alternative approach to residual stress manipulation has been explored in welding of high strength steels. Instead of adjusting welding parameters, the composition of the weld filler itself is changed to leverage the transformation strains associated with the austenite-to-martensite transformation. Ohta et al. investigated the residual stress variation in the welding of JIS SPV490 structural steel using a low transformation temperature (LTT) wire. They showed that adjusting the composition of the welding wire allowed to reduce the martensite start temperature to from ~500 °C to ~180 °C. Lower martensite start temperature enabled expansion of the weld metal in the final stage of solidification, inducing compressive residual stress within the weld. In contrast, in a conventional welding wire, tensile residual stresses were observed within the weldment. Such a change in the residual stress character had a profound effect in fatigue performance of the welded material, decreasing fatigue crack propagation rate throughout the investigated range of stress intensity factor. To date, use of LTT alloys in AM has not been investigated. However, Chen et al. demonstrated Ar arc cladding of LTT powder. They found high compressive residual stresses ranging from ~229 MPa to ~361 MPa within the clad coatings.

In this work, we adopt the concept of LTT materials in welding to demonstrate that solid-state phase transformations have crucial consequences for residual stress in metal AM parts. To investigate the effect of solid-state phase transformations, we compared two metals fabricated using DED: 1) pure Fe, and 2) Fe–50Cu (wt.%) binary alloy. The choice of these two materials was based on several considerations, discussed in detail below. First and foremost, Fe and Fe–50Cu undergo different phase transformations upon cooling from the melt. As shown in the Fe–Cu binary phase diagram and Fe cooling diagram, pure Fe solidifies first as body-centered cubic (BCC) Fe. It is then transformed into the face-centered cubic (FCC) austenite γ-Fe. Further cooling leads to a transition to BCC ferrite α-Fe. These BCC to FCC transitions are accompanied by a change in density and volume: density of δ-Fe at 1660 °C is 7.50 g/cm³, density of γ-Fe at 912 °C is 7.70 g/cm³, and density of α-Fe at 900 °C is 7.62 g/cm³. Consequently, the δ→γ transformation causes a ~3% decrease in volume, while the γ→α transformation causes a ~1% increase in volume. The phase transformation sequence of Fe–50Cu does not include δ-Fe: γ-Fe particles solidify directly within the liquid Cu-rich phase, and, upon cooling, the Fe-rich phase undergoes the γ→α transformation. This means that the phase transformation accompanied by the decrease in volume is eliminated in Fe–50Cu, while the phase transformation accompanied by an increase in volume is preserved. We can therefore hypothesize that this last γ→α transformation in Fe–50Cu will lead to formation of compressive residual stresses, similarly to the LTT welding wires discussed above, while parts fabricated from pure Fe will feature lower compressive or tensile fabricated stresses. Second, Fe–Cu alloys possess a unique combination of high electrical and thermal conductivity with high strength and good plasticity. However, a metastable miscibility gap that exists in the Fe–Cu system makes conventional processing (e.g., casting) of Fe–Cu alloys challenging. If the melt is undercooled into the metastable miscibility gap, it will decompose into two liquids, leading to formation of large spherical droplets of the minority phase within the majority phase. At low cooling rates, the minority phase droplets can aggregate, causing melt separation in Fe–Cu. As cooling rate
increases, solidification structures become more refined. AM processing of Fe–Cu alloys can enable finely dispersed microstructures due to the large cooling rates inherent to metal AM. However, to date, few studies report AM of Fe–Cu alloys. For example, Makarenko and Shishkovsky [62] demonstrated DED of multilayer functionally graded material (FGM) specimens fabricated from 316L stainless steel and aluminum bronze (10% Al). The SEM micrographs showed some evidence of liquid-phase separation with spherical Fe-rich particles of ~7 μm in diameter, as well as formation of Fe-rich dendrites and particles within the Cu-rich matrix.

In this study, we use DED to fabricate samples from pure Fe and Fe–50Cu (wt.%) binary alloy. By comparing residual stresses formed within the two alloys, we demonstrate that the solid-state transformation plays an important role in formation of residual stresses in AM-fabricated parts. Moreover, we show that DED is a feasible technique for fabrication of highly dense Fe–Cu parts with finely dispersed microstructures unattainable by conventional material processing approaches.

2. Materials and methods

Test samples (10 × 10 × 10 mm cubes) were fabricated using a laser engineered net shaping (LENS®) (Optomec, Albuquerque, NM, USA) 750 system equipped with a 1 kW 1064 nm YLR continuous wave laser (IPG Photonics, Oxford, MA, USA). The deposition chamber was kept in an inert Ar atmosphere, with oxygen level continuously monitored and maintained at <20 ppm. Samples were deposited onto an ASTM A36 low-carbon steel substrate plate 6.35 mm in thickness. Spherical gas-atomized powders of Fe and Fe–50Cu (wt.%) powders with nominal particle diameters of 38–150 μm were used as a feedstock material (TLS Technik GmbH & Co. Spezialpulver KG, Bitterfeld-Wolfen, Germany). A total of 18 samples (9 samples for Fe and 9 samples for Fe–50Cu) were fabricated to separate the effect of alloy composition and process parameters on the residual stresses. The process parameters that produce nearly fully dense samples were determined through a series of preliminary experiments as detailed below. Table 1 shows the directed energy deposition process parameters used in this study. For all samples, the laser thickness was set to 0.254 mm, hatch spacing was set to 0.4064 mm and working distance was set to ~13.7 mm to produce a laser spot 0.7 mm in diameter.

The porosity of printed samples was measured using optical microscopy (OM) with a BX53 M microscope (Olympus, Shinjuku, Tokyo, Japan). The images were processed using ImageJ software (NIH), and the total porosity was determined as the fraction of the porosity area to the total cross-sectional area. To color the ferrite grains, samples were etched using Klemm's I etch (1 g K₂S₂O₇ dissolved in 50 ml of stock solution of Na₂S₂O₃).

Residual stress measurements were performed by the hole drilling method according to the ASTM E837-13a standard by Hill Engineering (Rancho Cordova, CA, USA). A EA-06-031RE-120 type A strain gauge rosette (Vishay, Malvern, PA, USA) with three radial gauge elements was positioned on the top surface of the samples, and a total of 20 measurements were taken for depth profiling of residual stresses. Maximum and minimum principal residual stresses σ max and σ min were calculated as:

\[
\sigma_{\text{max}}, \sigma_{\text{min}} = \frac{\sigma_x + \sigma_y + \sqrt{(\sigma_x - \sigma_y)^2 + 4\tau_{xy}^2}}{2}
\]

where \(\sigma_x, \sigma_y\) – normal residual stresses (MPa), and \(\tau_{xy}\) – shear residual stress (MPa).

To distinguish the surface and bulk residual stresses, the top 0.5 mm of the samples through which the residual stress measurements were performed were divided into two zones: “surface” (<90 μm from the top surface) and “bulk” (>90 μm from the top surface). The residual stresses within the surface and bulk zones were then averaged, reducing the 20 measurements to 2 data points per sample. Analyses of variance (ANOVA) were performed on the bulk and surface datasets to determine the effect of the volumetric energy density (VED, J/mm³) and alloy composition on the total variation in residual stresses.

The elastic properties (i.e., Poisson’s ratio and Young’s modulus) were measured through the ultrasonic velocity technique with a 38DLP ultrasonic thickness gauge (Olympus, Shinjuku, Tokyo, Japan) equipped with a 5 MHz 0.25 in. diameter longitudinal wave contact transducer and a 5 MHz 0.25 in. diameter normal incidence shear wave contact transducer. Sample densities were measured using the Archimedes’ principle. Poisson’s ratio ν and Young’s modulus E were calculated as:

\[
\nu = \frac{1 - 2\frac{\rho}{\tau}}{2(1 + \nu)}
\]

\[
E = \frac{V_T\rho(1+\nu)(1-2\nu)}{1-\nu}
\]

where \(V_T\) – shear (transverse) velocity (mm/μs), \(V_L\) – longitudinal velocity (mm/μs), \(\rho\) – sample density (g/cm³).

Samples for microstructure examination were prepared by following the standard metallographic procedures. First, the samples were embedded in KuductoMet conductive phenolic mounting compound (Buehler, Lake Bluff, IL, USA). Then, the samples were polished with SIC papers (320–1200 grit) using an AutoMet automatic polisher (Buehler, Lake Bluff, IL, USA), followed by polishing with 3 and 1 μm diamond paste with extender on a canvas polishing surface. Finally, the samples were polished using aqueous alumina slurry on a micro-cloth pad in a GIGA-0900 vibratory polisher (PACE Technologies, Tucson, AZ, USA) for ~6 h.

Scanning electron microscopy (SEM) and energy-dispersive spectroscopy (EDS) were performed with a Magellan 400 XHR microscope (FEI, Hillsboro, OR, USA). Electron backscatter diffraction (EBSD) and focused ion beam (FIB) TEM sample preparation were performed using a GAIAD3 SEM-FIB dualbeam microscope (Tescan, Brno, Czech Republic) equipped with a NordlysMax2 EBSD detector (Oxford Instruments, Abingdon, Oxfordshire, UK) and a Ga ion source. EBSD maps were post-processed using the open source MTEX Matlab toolbox [40]. Data points with mean angular deviation (MAD) of >1° were removed, and grain reconstruction was performed by defining grains as areas of the map completely surrounded by boundaries with a misorientation >5. The orientation data were denoised and missing data were interpolated.

<table>
<thead>
<tr>
<th>Sample ID</th>
<th>Material</th>
<th>Laser power (W)</th>
<th>Scan speed (mm/s)</th>
<th>Powder feed rate (g/s)</th>
<th>Specific energy (J/mm³)</th>
<th>VED (J/mm³)</th>
<th>Powder mass density (g/mm³)</th>
</tr>
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<tr>
<td>60-62</td>
<td>Fe</td>
<td>467</td>
<td>26.67</td>
<td>0.365</td>
<td>25</td>
<td>100</td>
<td>0.020</td>
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<tr>
<td>63-65</td>
<td>Fe</td>
<td>889</td>
<td>16.93</td>
<td>0.237</td>
<td>75</td>
<td>300</td>
<td>0.020</td>
</tr>
<tr>
<td>66-68</td>
<td>Fe</td>
<td>926</td>
<td>10.58</td>
<td>0.170</td>
<td>125</td>
<td>500</td>
<td>0.023</td>
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<tr>
<td>73-75</td>
<td>Fe-50Cu</td>
<td>889</td>
<td>16.93</td>
<td>0.271</td>
<td>75</td>
<td>300</td>
<td>0.023</td>
</tr>
<tr>
<td>101-103</td>
<td>Fe-50Cu</td>
<td>400</td>
<td>16.09</td>
<td>0.270</td>
<td>36</td>
<td>140</td>
<td>0.024</td>
</tr>
<tr>
<td>104-106</td>
<td>Fe-50Cu</td>
<td>380</td>
<td>6.35</td>
<td>0.245</td>
<td>85</td>
<td>340</td>
<td>0.055</td>
</tr>
<tr>
<td>107-109</td>
<td>Fe-50Cu</td>
<td>450</td>
<td>16.93</td>
<td>0.247</td>
<td>38</td>
<td>152</td>
<td>0.021</td>
</tr>
</tbody>
</table>

* VED – volumetric energy density.
using the half-quadratic filter [41]. An approach based on local orientation changes was employed to calculate the density of geometrically necessary dislocations (GNDs) [42]. Scanning transmission electron microscopy (STEM) was performed with a JEM-2800 S/TEM microscope outfitted with dual 100 mm² silicon drift detectors (SDD) for EDS.

3. Results

3.1. Powder characterization

First, we investigated the morphology and composition of the gas atomized Fe and Fe–50Cu powders (Fig. 1). The Fe powder particles were found to exhibit spherical shape (Fig. 1a), while the Fe–50Cu powder featured more irregularly shaped particles (Fig. 1b). Powder diffraction patterns collected from the Fe and Fe–50Cu samples confirmed that no extraneous phases were present in the feedstock powders (Fig. 1c).

3.2. DED process parameters optimization

To determine the optimal processing parameters for LENS® of pure Fe parts, we deposited multiple 10 × 10 × 10 mm cuboid test samples. We used specific energy \( E (J/mm^2) \) and powder mass density \( G (g/mm^2) \) as combined parameters guiding the optimization:

\[
E = \frac{P}{2v_r} \quad (4)
\]

\[
G = \frac{\dot{m}}{2v_r} \quad (5)
\]

where \( P \) is the laser power (W), \( v \) is the scan speed (mm/s), \( r_l \) is the radius of the laser spot and \( \dot{m} \) is the feed rate of the powder (g/min, \( 1.67 \times 10^{-5} \) kg/s).

Fig. 2 shows that at high \( E/G \) ratios, the dilution problem is present, i.e., not enough powder is supplied to successfully build the parts. If the \( E/G \) ratio is low, the porosity problem is evident, i.e., the laser power is not high enough to melt all the particles, leading to formation of lack-of-fusion porosity. The feasible processing window is indicated in Fig. 2 with a green oval.

Additionally, the effect of specific energy on the porosity of the parts is illustrated in Fig. 3. Lower specific energy yielded highly porous samples with both lack-of-fusion and gas porosity present. Higher specific energy produced parts with a decreased amount of lack-of-fusion and gas porosity.

3.3. Elastic properties

The measured Poisson’s ratio and Young’s modulus for the two additively manufactured materials, Fe and Fe–50Cu, are presented in Table 2. Poisson’s ratio was determined to be 0.28 and 0.33 for Fe and Fe–50Cu, respectively. Young’s modulus was calculated as \(~201\) GPa for Fe and \(~144\) GPa for Fe–50Cu. These values are consistent with the literature data (\(~208\) GPa for Fe) and the rule of mixtures, through which the estimated modulus of Fe–50Cu is estimated to be \(~168\) GPa [151].

3.4. Microstructure of Fe DED deposits

Optical micrographs of the microstructure of a representative pure Fe LENS® build are shown in Fig. 4a. At the bottom of the build, the heat affected zone between the low-carbon steel substrate and the pure Fe build is present. The pure Fe part exhibits grains elongated along the build direction, as is typical for AM. Fig. 4b presents the microstructural variations along the build direction. White arrows indicate gas porosity, while no lack-of-fusion porosity was observed in the builds fabricated using the optimized parameters.

Fig. 5 presents SEM micrographs of a representative Fe sample fabricated at a VED of 100 J/mm³. Melt pool boundaries are visible from the low magnification micrographs (Fig. 5a), and no lack-of-fusion porosity is observed within the sample. In the higher magnification micrographs (Fig. 5b), a tendency for spherical gas pores of \(~230\) nm in diameter to migrate to the grain boundaries is evident. The pores then coalesced, promoting hot cracking at the grain boundaries [63].
Fig. 3. Effect of specific energy on porosity of Fe parts printed via directed energy deposition (DED). Higher specific energy used during DED reduces the amount of lack of fusion porosity.

Table 2
Material elastic constants determined from the ultrasonic velocity measurements.

<table>
<thead>
<tr>
<th>Material</th>
<th>Shear velocity (mm/μs)</th>
<th>Longitudinal velocity (mm/μs)</th>
<th>Density (g/cm³)</th>
<th>Poisson’s ratio</th>
<th>Young’s modulus (GPa)</th>
<th>Shear Modulus (GPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fe</td>
<td>3.193</td>
<td>5.788</td>
<td>7.69</td>
<td>0.28</td>
<td>201</td>
<td>78</td>
</tr>
<tr>
<td>Fe-50Cu</td>
<td>2.624</td>
<td>5.231</td>
<td>7.85</td>
<td>0.33</td>
<td>144</td>
<td>54</td>
</tr>
</tbody>
</table>

Fig. 4. Optical micrographs showing the microstructure of a representative pure Fe LENS® build (scale bar = 200 μm). a) An optical micrograph of an entire Fe LENS® build. b) Microstructural variations along the build direction. White arrows indicate gas porosity.
3.5. Microstructure of Fe–50Cu DED deposits

SEM micrographs of a representative microstructure of a Fe–50Cu deposit are shown in Fig. 7. In these secondary electron micrographs, the Fe-rich phase appears darker than the Cu-rich phase due to a lower atomic number of Fe as compared to Cu. The Fe-rich phase forms finely distributed, nearly spherical particles or elongated dendrites, depending on the position within the melt pool.

EDS analysis revealed that dendrites and spherical particles are Fe-rich, while inter-dendritic regions are Cu-rich (Fig. 8). After LENS® processing, composition of the Fe–50Cu alloy remained close to that of the pristine powder: 49.1–49.8 wt% Fe and 50.2–50.9 wt% Cu (Table 3). This result indicates that Cu loss during DED processing was minimal, despite its significantly lower melting point (1538 °C and 1085 °C for Fe and Cu, respectively [64,65]).
3.6. Residual stresses in DED-printed samples

Results of residual stress measurements are presented in Fig. 9. For samples fabricated from pure Fe feedstock powder, higher energy input during printing yielded residual stresses lower in magnitude and of a more compressive character (Fig. 9a). For Fe–50Cu samples, surface compressive stresses of ~190 MPa were observed (Fig. 9b). High variability was observed between the replicate samples fabricated with the same process parameters.

The mean values of maximum principal residual stress in the bulk and surface zones of the Fe and Fe–50Cu samples as a function of VED are plotted in Fig. 10. For pure Fe samples, higher VED used during DED caused lower stress both in the bulk and on the surface of the samples. For Fe–50Cu, higher VED caused higher bulk stresses and did not affect the surface stresses. This result suggests that the relationship between the residual stress and process parameters is material-specific, corroborating the findings of Denlinger et al. obtained for Ti–6Al–4V and Inconel 625 [43] and validating our initial hypothesis.

The ANOVA results are presented in Table 4. For the surface residual stresses, there was a statistically significant difference between the Fe and Fe–50Cu samples as determined by one-way ANOVA ($F(1,5) = 80.181, p = 1.16 \times 10^{-6}$). Moreover, the difference between the samples fabricated with different VED values was also statistically significant ($F(1,5) = 7.977, p = 0.00223$). Based on the ANOVA results, ~64.6% of the variation in surface principal residual stresses was attributed to the alloy composition, ~25.7% to the VED, and ~9.7% of the variation was not explained by the statistical model and was attributed to random error. For the bulk residual stresses, both VED and material factors had a statistically significant effect on the bulk principal residual stresses ($p = 3.93 \times 10^{-6}$ and $p = 2.14 \times 10^{-5}$, respectively). Variation partitioning based on the ANOVA results suggested that ~25.7% of the variation in bulk principal residual stress was attributed to the alloy composition, ~67.4% - to the VED, and ~6.9% - to the random error. The statistical analysis results confirmed our initial hypothesis that the residual stresses in additively manufactured metal parts can be manipulated by adjusting the alloy composition. Moreover, adjusting the process parameters also had a statistically significant effect on both bulk and surface residual stresses.

3.7. Plastic deformation in DED-printed Fe–50Cu

Phase-specific plastic deformation within the Cu-rich and the Fe-rich phases of DED-fabricated Fe–50Cu alloy was examined directly with TEM. A TEM sample was extracted with a focused ion beam lift out procedure from within the top 500 μm of the LENS® deposit (Fig. 11a). Examination of the sample using TEM and STEM revealed a high density of dislocations within the Cu-rich phase (Fig. 11b and c), and dislocation-free Fe-rich dendrites and particles. This was further confirmed with STEM-EDS, which demonstrated conclusively a high dislocation density within the Cu-rich phase (Fig. 11d).

To quantify the GND density within the two phases, we performed EBSD mapping of a Fe–50Cu sample (Fig. 12). A phase map showing the distribution of the Fe-rich and Cu-rich phases is presented in Fig. 12a. The inverse pole figure (IPF) map of the Fe-rich phase (Fig. 12b) demonstrates that each of the Fe-rich particles and dendrites is a separate grain. In contrast, the IPF map of the Cu-rich phase (Fig. 12c) suggests gradual orientation changes within the Cu-rich phase and formation of relatively large, micro-scale grains. Phase-specific analysis of the GND density revealed that GND densities were log-normally distributed within both phases (Fig. 12d). The mean GND density was calculated to be $471 \pm 95 \, \mu \text{m}^{-2}$ in the Fe-rich phase and $684 \pm 80 \, \mu \text{m}^{-2}$ within the Cu-rich phase, and a t-test performed on the difference in means confirmed statistical significance of the difference in GND densities.

Table 3

<table>
<thead>
<tr>
<th>Site</th>
<th>Element</th>
<th>Wt.%</th>
<th>At.%</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Fe</td>
<td>49.8</td>
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<tr>
<td></td>
<td>Cu</td>
<td>50.2</td>
<td>47.0</td>
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<tr>
<td>2</td>
<td>Fe</td>
<td>49.3</td>
<td>52.5</td>
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<tr>
<td></td>
<td>Cu</td>
<td>50.7</td>
<td>47.5</td>
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<tr>
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<td>Cu</td>
<td>50.9</td>
<td>47.6</td>
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</table>

Fig. 8. Energy dispersive spectroscopy mapping of a representative Fe–50Cu deposit showing the distribution of the Fe-rich and Cu-rich phases.
4. Discussion

DED-printed parts of pure Fe tended to exhibit large grains elongated along the build direction (Figs. 4, 5, Fig. 6), which is a microstructure typical for additively manufactured parts. Fe–Cu50 parts, in turn, showed a fine dispersion of the Fe-rich phase within the Cu-rich matrix and small, randomly oriented grains (Figs. 8 and 12). Interestingly, such finely-dispersed microstructures cannot be readily achieved in immiscible alloys in general and Fe–Cu binary alloys specifically using between the two phases (p = 0.0008).

Fig. 9. Residual stress measurement results. Data for individual samples are shown in grey traces. Red (for Fe) and blue (for Fe–50Cu) traces show mean values of residual stresses calculated for three replicate samples. Error bars represent 95% confidence intervals on the mean. a) Maximum principal residual stress in Fe samples fabricated at three VED values: 100 J/mm³ (left), 300 J/mm³ (center) and 500 J/mm³ (right). b) Maximum principal residual stress in Fe–50Cu samples fabricated at three VED values: 140 J/mm³ (left), 300 J/mm³ (center), and 340 J/mm³ (right). (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

Fig. 10. Maximum principal residual stress in the (a) bulk and (b) surface zones of the Fe and Fe–50Cu samples as a function of volumetric energy density (VED).

Table 4
Analysis of variance results for bulk and surface residual stresses.

<table>
<thead>
<tr>
<th>Source</th>
<th>DF</th>
<th>SS</th>
<th>MS</th>
<th>F-value</th>
<th>p</th>
</tr>
</thead>
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<td><strong>Surface residual stress</strong></td>
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<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Material</td>
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<td>132199</td>
<td>80.181</td>
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<tr>
<td>VED</td>
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<td>52607</td>
<td>13152</td>
<td>7.977</td>
<td>0.00223</td>
</tr>
<tr>
<td>Residuals (error)</td>
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<td>19785</td>
<td>1649</td>
<td></td>
<td></td>
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<tr>
<td><strong>Bulk residual stress</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Material</td>
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<td>12470</td>
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<td>2.14×10⁻⁵</td>
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<tr>
<td>VED</td>
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<td>29.57</td>
<td>3.93×10⁻⁶</td>
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<td>Residuals (error)</td>
<td>14</td>
<td>3314</td>
<td>276</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

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conventional manufacturing methods, such as casting [64,65].

Hole drilling residual stress measurements demonstrated that both the magnitude and the character (i.e., tensile/compressive) of the residual stress can be manipulated by engineering solid-state transformations in the alloy. By separating the stresses into surface and bulk components and applying ANOVA to the resulting data, we were able to demonstrate that surface residual stresses are controlled by the alloy composition. For surface residual stress, ~65% of the total variation was attributed to the alloy composition, ~25% to DED process parameters summarized in VED, and ~10% to other factors and random error. In contrast, for bulk residual stress, the variation was dominated by DED process parameters: ~67% of the variation was attributed to VED, ~26% to alloy composition, and 7% to random error.

It is important to note that the present analysis of the residual stresses is limited by the hole-drilling technique, in which only in-plane stresses can be measured. No information about the build direction stresses can be acquired with a strain gauge placed on the top surface of the sample. Future studies will involve additional strain gauges placed on side surfaces of the sample or other residual stress measurement approaches that are capable of measuring stresses in x, y and z directions, such as neutron diffraction.

Overall, residual stresses were more compressive in the Fe–50Cu samples. We can speculate that this effect can be attributed to the phase transformations occurring during solidification of the Fe–50Cu binary alloy. According to the Fe–Cu binary phase diagram, the FCC γ-Fe phase solidifies first, followed by solidification of the ε-Cu phase and a solid-state phase transformation of FCC γ-Fe to BCC α-Fe. Upon further cooling, the system stabilizes as a two-phase solid with the Cu-rich phase as a matrix. For reference, Fig. 13a shows the Fe–Cu binary phase diagram, as well as the corresponding changes in crystal structure and lattice strain, see Fig. 13b.

A schematic representation of the phase and microstructure evolution during solidification of Fe–50Cu binary alloy is shown in Fig. 14. Solid-state phase transformation of FCC γ-Fe to BCC α-Fe is accompanied by a ~1% volume increase of the Fe-rich particles/dendrites due to the BCC lattice accommodating fewer atoms in a unit cell than the FCC lattice.

We can now discuss the phase and microstructure evolution in terms

Fig. 11. Scanning transmission electron microscopy (STEM) imaging of a DED-fabricated Fe–50Cu sample. a) An SEM micrograph of the TEM sample. b–c) Bright-field TEM and STEM micrographs, respectively, showing high dislocation density within the Cu-rich phase. d) STEM-EDS mapping confirming that dislocations are confined to the Cu-rich phase.

Fig. 12. Electron backscatter diffraction (EBSD) mapping of a DED-fabricated Fe–50Cu sample. a) Phase map showing the distribution of the Fe-rich and Cu-rich phases. b) Inverse pole figure (IPF) map of the Fe-rich phase. c) IPF of the Cu-rich phase. d) Histogram of the geometrically necessary dislocation (GND) density for the two phases, with mean GND densities plotted in the inset. Error bars indicate 95% confidence intervals.
of the residual stresses within the system upon cooling (see Fig. 15). Let us first focus on pure Fe. In the liquid state, residual stresses within this system are equal to zero. Upon cooling, the BCC $\delta$-Fe phase solidifies first. Cooling of $\delta$-Fe results in thermal contraction of the sample that is constrained by the substrate, which leads to development of hydrostatic tensile residual stresses near surfaces, balanced by compressive stresses in the bulk. At $\sim1394^\circ C$, the BCC $\delta$-Fe undergoes a solid-state phase transformation to FCC $\gamma$-Fe accompanied by $\sim3\%$ decrease in volume. The decrease in volume is associated with a rapid increase in surface tensile residual stress. After the phase transformation is complete, the system once again undergoes only thermal contraction, which continues to increase residual stresses until the system reaches $\sim910^\circ C$. At this temperature, FCC $\gamma$-Fe is transformed to BCC $\alpha$-Fe, causing a $\sim1\%$ increase in volume, which slightly decreases the residual stress. Finally, upon further cooling, the surface tensile residual stresses increase due to thermal contraction of $\alpha$-Fe.

Fig. 13. (a) Fe–Cu binary phase diagram, and (b) phase transformations and increases in volume, adapted from Ref. [57].

Fig. 14. A schematic representation of phase evolution during solidification of the Fe–50Cu alloy.

Fig. 15. A schematic of the evolution of residual stress during solidification and cooling of pure Fe and the Fe–50Cu alloy.
The evolution of residual stresses in the Fe–50Cu alloy follows a different path. First, we can consider the Fe-rich phase. The γ-Fe particles and dendrites solidified within the liquid Cu-rich phase will be virtually stress-free. Upon solidification of ε-Cu that starts at ~1085 °C, thermal contraction of the Cu-rich matrix induces compressive residual stresses within the γ-Fe particles and dendrites. At ~910 °C, solid-state phase transformation of γ-Fe to α-Fe (accompanied by volume expansion of the Fe particles) leads to an increase in compressive residual stresses. Further cooling of the system leads to thermal contraction of the particles and, consequently, to a decrease in compressive residual stresses. The Cu-rich phase follows a similar path. Liquid Cu-rich phase remains stress-free until ~1085 °C, at which point solidification, cooling and resulting thermal contraction of Cu results in hydrostatic tensile residual stresses within the matrix. At ~910 °C, expansion of the Fe-rich particles caused by the solid-state transformation of Fe introduces compressive residual stresses. Finally, cooling of the system results in contraction of both α-Fe and ε-Cu that causes a decrease in the magnitude of compressive stress within the Cu-rich phase.

If we contrast the residual stress evolution within the pure Fe to that of Fe–50Cu, we can clearly see that, upon cooling, residual stresses in pure Fe are likely to remain tensile. However, both Cu-rich and Fe-rich phases within Fe–50Cu are likely to exhibit compressive residual stresses. It is important to note that the yield strength of Cu is significantly lower than that of Fe (69 MPa [66] versus 140 MPa [67], respectively), which explains our observations of high dislocation density within the Cu-rich phase (Figs. 11 and 12).

While we can attribute the observed differences in residual stress state to the solid-state phase transformations within the Fe and Fe–50Cu samples, it is important to highlight that DED is a non-equilibrium, high cooling rate processing technique, in which phase transformations might deviate from those predicted by the equilibrium phase diagram. An in situ synchrotron diffraction study of Fe–50Cu DED could be used to acquire real-time information on the phases and strains present within the material.

5. Conclusions

In this work, we investigated the relationship between alloy composition and residual stresses formed during DED using a model binary system consisting of both hard and soft metallic phases. To this end, we studied DED-fabricated components of: i) pure Fe, and ii) Fe–50Cu (wt.%) binary alloy. We measured the residual stresses within as-deposited samples using the hole drilling approach and applied ANOVA statistics to analyze the results. TEM and EBSD characterization of the Fe–50Cu builds revealed a high density of dislocations within the Cu-rich phase, suggesting that plastic deformation in the Cu-rich phase can accommodate the residual stresses introduced during DED. These findings, coupled with an analysis of the complex phase transformations in Fe-based alloys, demonstrate that phase transformations and alloy compositions can be successfully engineered to produce a desired residual stress state in as-deposited DED components.

References

