Tuning Microstructure and Mechanical Properties of Additively Manufactured CoCrFeMnNi High Entropy Alloy by Deep Cryogenic Treatment

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Abstract: Here, deep cryogenic treatment (DCT) is proposed to be an easy and effective approach to tune the microstructure and thus enhance the mechanical performance at room temperature of the laser melting deposited CoCrFeMnNi high entropy alloy (HEA). DCT is found to induce large compressive residual stress and crystalline defects including dislocations, stacking faults, nanotwins and nanograins into the additively manufactured CoCrFeMnNi HEA. Nano-twinning plays a critical role in the strengthening, contributing 51.0–246.8 MPa to the DCT-processed CoCrFeMnNi HEA, while the compressive residual stress is thought to be responsible for the improved ductility by suppressing crack formation. The crystalline defects are evidently generated during the cryogenic soaking, which also result in the compressive residual stress upon reheating at the end of the DCT. The present work establishes DCT as an effective method to simultaneously enhance the strength and plasticity in alloys fabricated by additive manufacturing.

Keywords: High entropy alloy, Deep cryogenic treatment, Laser directed energy deposition

1 Introduction

Laser directed energy deposition (LDED) has been established as a versatile additive manufacturing (AM) technology for the direct fabrication of complex shaped alloy products from powder feedstocks. However, repeated melting and cooling occurred in a molten pool during LDED can lead to a high temperature gradient and thus unevenly distributed residual stress within the as-built components, which will inevitably influence the mechanical performance of the AM components, especially their strength and ductility. DCT has recently been proposed as a nondestructive method for effectively tuning the residual stress state and microstructure for improving the mechanical properties of Ti and Ni alloys such as the TC6 alloy, IN718 super-alloy, and Ti-6Al-4V alloy. In this work, using the CoCrFeMnNi HEA as a prototypic alloy system, we demonstrate that DCT can produce beneficial effects in LDED-fabricated samples. The influence of DCT on the residual stress, microstructural evolution, and mechanical properties of LDED-fabricated CoCrFeMnNi HEA was elucidated. Specific emphasis is placed on revealing the formation of deformation microstructures at cryogenic temperature via microscopy observation to understand the contributions from different strengthening mechanisms to the yield strength of the DCT-processed CoCrFeMnNi HEA samples, and using finite element modeling (FEM) to quantitatively rationalize the formation of the residual stress.

2 Experimental procedure

All the HEA samples were fabricated using an additive manufacturing system. The feedstock material here is the spherical or near spherical gas-atomized CoCrFeMnNi HEA powders. After LDED, all of the samples were cooled to room temperature in the operating chamber, and DCT were immediately carried out to cool the LDED-built samples in the liquid nitrogen chamber. When the samples reached 77 K, they were soaked at this temperature for different durations. The samples subjected to DCT by soaking for 12 h, 24 h, 48 h and 120 h at liquid nitrogen temperature are hereafter labeled as DCT12h, DCT24h, DCT48h and DCT120h, respectively. Details of the DCT process are shown in Fig. 1.

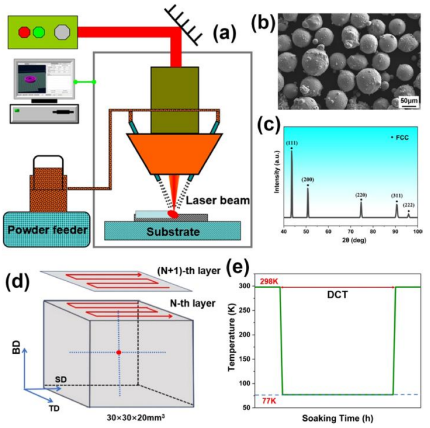

Fig. 1. LDED of CoCrFeMnNi HEA: (a) LDED experimental setup, (b) and (c) SEM image and XRD pattern of the raw powders, and (d) the scanning strategy during the LDED process, and (e) DCT process.

The microstructure was observed by using SEM equipped with EBSD, and TEM with EDS. Room temperature tensile tests were carried out using a universal testing machine at 10^3 s^{-1}. A nonlinear FEM model was established using the commercial software ABAQUS for the coupled thermo-mechanical simulation of the LDED process, and the stress

development during the DCT process was simulated using a visco-elastic-plastic model.

3 Result and discussion

All the XRD spectra display only a typical FCC structure, consistent with that of the raw powders, indicating that no new phases were formed and the FCC structure did not change during the DCT process. The compressive residual stress shows a gradual rising trend with the soaking duration, with the DCT120h sample exhibiting the highest compressive residual stress of ~415 MPa measured among all the samples. The variation trend of residual stress in the DCT-processed samples is opposite to that of the LDED-built samples treated via annealing.

The microstructure of the as-built CoCrFeMnNi HEA sample consists mainly of dominant columnar grains, while fine equiaxed grains appear only near the fusion lines indicated between the adjacent deposited layers. There is a high density of dislocations in the as-built sample, which is attributed to the high thermal stress and accumulated micro strain caused by a complex thermal history of the LDED process. Interestingly, in addition to a high density of dislocations located in the matrix, striped nanotwins are observed in the form of bundles in the DCT-processed samples. When the soaking duration is increased to 120 h, a high-density of dislocations, stacking faults and nanotwins exist in the matrix of sample. Homogeneously distributed nanograins (grain size of ~25 nm) can be observed in local regions of the DCT120h sample.

We have conducted the uniaxial tensile tests at ambient temperature and the representative engineering stress-strain curves are plotted in Fig. 2(a). The as-built CoCrFeMnNi HEA possesses a yield strength (σ_y) of 290 ± 9 MPa, ultimate tensile strength (σ_{UTS}) of 456 ± 17 MPa and elongation to failure (ε_f) of 34.8 ± 2.1%. After subjected to DCT for different soaking durations, all of the samples exhibit higher σ_y, σ_{UTS} and ε_f values than the as-built sample, indicating that DCT can promote a significant improvement in both strength and ductility.

How are the deformation microstructures responsible for the strengthening of the DCT-processed samples produced during the DCT process? In this connection, the following should be noted:

(I) The crystalline defects that contribute to strengthening are evidently produced during the cryogenic soaking of the DCT process. The nanotwin density increases on prolonging the DCT duration, and strengthening due to dislocation density and twinning also increases with the prolonged DCT duration.

(II) A state of compressive residual stress is also observed from the DCT-processed samples, with increasing magnitude on increasing DCT duration.

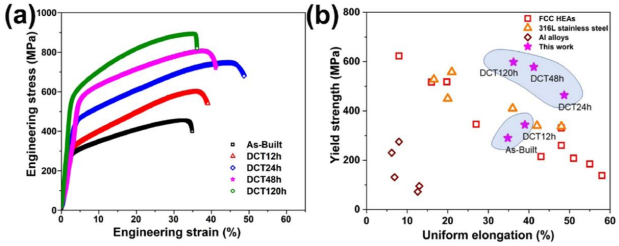

Fig. 2(a) Engineering stress-strain curves of the as-built and DCT-processed HEA samples. (b) Comparison of the yield strength (σ_y) and elongation to failure (ε_f) obtained in this work with FCC HEAs fabricated by various preparation and post-processing methods.

Conclusion

1. Both the as-built and DCT-processed CoCrFeMnNi HEA samples exhibit a single FCC phase, demonstrating their excellent phase stability.

2. DCT causes compressive residual stresses and introduces many defects composed of dislocations, stacking faults, deformation nanotwins and nanograins into the as-built samples, which lead to a significant enhancement in both strength and plasticity.

3. The defects are evidently formed during the prolonged cryogenic soaking of the DCT process under the thermal stress state due to the initial cryogenic cooling.

Acknowledgments

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