

ORIGINAL RESEARCH ARTICLE

Cracking mechanism in single-pass molten pool of laser-directed energy deposition-manufactured FeCoCrNiAl high-entropy alloy coatings

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Abstract

To meet the critical strategic demands of deep-sea and polar exploration, there is an urgent need to develop a new generation of marine exploration equipment with enhanced corrosion resistance, drag reduction, and ice prevention capabilities. High-entropy alloy (HEA) coatings are an effective way to address these problems, offering excellent corrosion resistance, high mechanical properties, and drag-reduction characteristics. Laser-directed energy deposition (LDED) additive manufacturing is an advanced manufacturing technique for fabricating HEA coatings, enabling the deposition of components on complex curved surfaces while delivering excellent overall performance. However, the inherent cracking problem in LDED-processed HEAs accelerates rapid damage and coating failure. In this work, the FeCoCrNiAl HEA coatings were fabricated on the GCr18Mo substrate using LDED. The crack types and underlying cracking mechanisms, as well as crack-induced hardness degradation mechanisms, are elucidated. The results show that the defect types include solidification cracks, heat-affected zone (HAZ) cracks, and gas pores, with solidification cracks accounting for a substantial proportion. The hot stress in the molten pool exceeds the coating strength. The residual tensile stress (~900 MPa) in the HAZ exceeds the yield strength of the matrix, while stress fluctuations in the molten pool during solidification triggered crack initiation. The cracking mechanism is closely related to grain orientation, dislocation accumulation, and carbide. The geometrically necessary dislocation density at the crack ($\sim 1.33 \times 10^{14} \text{ m}^{-2}$) is higher than that in the matrix. Hard carbide particles play a critical role in crack initiation. Moreover, solidification cracks are a mixture of trans-granular and inter-granular cracks. The hardness of the molten pool, HAZ, and interface zones is higher than that of the substrate, with the HAZ exhibiting a hardness 3.53 times higher. Cracks cause a localized reduction in hardness, with the most pronounced decrease occurring in the solidification crack region. Clarifying the cracking mechanism of the LDED-manufactured HEA coatings will contribute to breakthroughs in marine engineering equipment.

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1. Introduction

To meet the critical strategic demands of deep-sea and polar exploration, where marine structures are subjected to harsh service environments characterized by low temperatures, high humidity, and severe corrosion, there is an urgent need to develop a new generation of marine exploration equipment that can overcome key technological bottlenecks in marine engineering, with properties such as corrosion resistance, drag reduction, and ice prevention.^{1,2} Protective coatings are among the most effective strategies for drag reduction and ice prevention in marine engineering and equipment, with metallic coatings being particularly prominent.³ Bare GCr18Mo used directly as a substrate in marine environments exhibits inherent limitations due to its insufficient corrosion resistance. In particular, when exposed to harsh marine conditions, uncoated GCr18Mo is highly susceptible to localized corrosion, especially pitting corrosion, which can significantly deteriorate its service performance.⁴ In addition, although some surface modification methods can provide reinforced coatings, their thickness is typically limited (on the order of $\sim 20\ \mu\text{m}$), which may not be sufficient to ensure long-term protection under severe marine conditions.⁵ Laser-directed energy deposition (LDED) is a key technology for the green, low-carbon, and efficient fabrication of metal coatings. A high-energy laser beam simultaneously melts the matrix surface and the supplied metal powder, followed by rapid solidification to produce high-quality metal coatings.^{6,7} Its main advantages include high repair efficiency, strong coating interface adhesion, and excellent repair quality.^{6,8}

High-entropy alloy (HEA) coatings represent a breakthrough in marine engineering, attracting significant attention due to their excellent corrosion resistance and superior drag reduction properties.^{9,10} HEA coatings inherit the “four core effects” of bulk HEAs, especially high entropy and slow diffusion, which enable high-quality coatings to exhibit excellent service characteristics.¹¹ Recently, the LDED additively manufactured HEA coatings have attracted much attention, with some research progress achieved and applications found in marine engineering.^{12,13} Wang *et al.*¹⁴ successfully developed corrosion-resistant LDED-manufactured FeCoNiCrMo_x HEA coatings, achieving improved inter-granular corrosion resistance in the passivated state. Zhang *et al.*¹⁵ studied the successful design and fabrication of the LDED-manufactured functionally graded HEA coatings on a marine EH36 substrate. Xiao *et al.*¹⁶ analyzed the effects of dilution on

the microstructure evolution and corrosion behavior of LDED-manufactured FeCoNiCrNb_{0.5} coatings on marine engineering equipment. The high dilution ratio and severe segregation reduce the appearance of the eutectic structure, which cannot inhibit the expansion of pitting corrosion and reduce the corrosion resistance. Thus, the LDED-manufactured HEA coatings in high-end manufacturing fields such as ship repair have excellent hardness, strength, and impact resistance, which play a key role under extreme temperature and complex stress conditions.

However, the HEA coatings fabricated by LDED suffer from inherent defects (cracks, pores, etc.), exacerbating corrosion and mechanical failure.^{17,18} Mooraj *et al.*¹⁹ investigated solid-state cracking in the LDED-fabricated AlCrFe₂Ni₂ HEAs. Solid-state cracking occurred in the single-phase body-centered cubic (BCC)/B2 microstructure, accompanied by large residual strain, which limited the mechanical properties of the HEA coatings. Cui *et al.*²⁰ studied the processing defects and damage mechanisms in the LDED-manufactured Ti₄₁V₂₇Hf₁₃Nb₁₃Mo₆ refractory HEAs, and lack-of-fusion defects act as primary crack-initiation sites, promoting brittle fracture under tensile loading. Besides, cracking dominates in moderate-defect refractory HEAs, whereas pore nucleation dominates in low-defect ones. Zhang *et al.*²¹ found significant crack defects in the LDED-manufactured CoCrFeNiTi_x HEA coatings, predominantly in the form of longitudinal cracks, which exhibit characteristics of trans-granular fracture. The cracking mechanisms in the LDED-manufactured HEA coatings are essential for the development of high-performance coatings. Recently, several researchers have investigated cracking in the LDED-fabricated coatings. Zeng *et al.*²² investigated the formation mechanism of hot cracks in LDED-fabricated multi-pass and multi-layer DD6 single-crystal superalloys. Non-directional dendritic growth generates impurity segregation that promotes the initiation and propagation of hot cracks. Furthermore, the locations and propagation directions of cracks depend on regions of stress-strain concentration. Li *et al.*²³ analyzed crack behavior of the LDED-manufactured GH4169/K477 heterostructure HEA coatings. A low-melting-point eutectic in the heat-affected zone (HAZ) is a potential source of liquid film and leads to the initiation of liquefaction cracks. Ouyang *et al.*²⁴ investigated the micro-crack initiation mechanism of HEA alloys manufactured by LDED. Hot stress can readily rupture fragile grain boundaries, resulting in

the formation of inter-granular cracks. Therefore, the cracking mechanism of the LDED-manufactured metal coatings is closely related to thermal stress, grain size, and precipitates. However, existing studies on cracking in LDED-processed materials generally focus only on one or several isolated mechanisms, such as thermal stress, grain-boundary weakening, elemental segregation, or precipitate-induced cracking, rather than providing a systematic and unified explanation of crack initiation and propagation behavior. In addition, relatively limited work has been conducted on FeCoCrNiAl HEAs from the perspective of a comprehensive crack evolution framework under LDED conditions.

In this work, taking the LDED-manufactured FeCoCrNiAl HEAs coating as the research object, the cracking mechanism of the coating is studied by combining simulation and experiments. The relationship between LDED manufacturing parameters and crack defects is established. The residual equivalent, tensile, and compressive stress evolution during the LDED solidification process is calculated. The grain, precipitate, and dislocation characteristics at the crack region are analyzed. The hardness distribution across the coating, interface, HAZ, and matrix is measured. This work clarifies the mechanisms of crack initiation and propagation, as well as the correlation between cracking behavior and hardness variation. Importantly, although the experimental study is based on a single-pass molten pool, it represents the fundamental building block of multi-track LDED deposition. The intrinsic thermo-mechanical behavior, including thermal gradient-driven stress evolution and solidification cracking tendency, is governed by local melt pool physics, which remains applicable to multi-track and multi-layer conditions at the fundamental scale. Therefore, the present results provide essential mechanistic insight for understanding crack formation in practical LDED coating systems.

2. Materials and methods

2.1. Materials and laser-directed energy deposition process

The GCr18Mo steel was used as the substrate, with

dimensions of 150 mm × 120 mm × 10 mm. Cr18Mo steel is a bearing steel grade defined under the Chinese National standard system and does not have a strictly equivalent American Society for Testing and Materials designation. However, it belongs to the high-carbon chromium bearing steel family and is compositionally comparable to American Society for Testing and Materials A295 Grade 52100, which is widely used as a reference material for similar applications. The primary difference lies in the addition of molybdenum in GCr18Mo, which enhances hardenability and performance. Table 1 shows the chemical composition of the FeCoCrNiAl HEA powder and matrix. FeCoCrNiAl HEA powder served as the LDED material. The particle morphology and size distribution are presented in Figure 1a and 1b. The powder has an equiatomic composition of iron, copper, chromium, nickel, and aluminum. The elemental content of the powder is shown in Figure 1c.

All samples were produced with the LDED system (iLAM, Nanjing HuiRui optoelectronic technology Co., Ltd., China) shown in Figure 1d, consisting of a 6,000 W continuous fiber laser, coaxial powder feeder, shielding gas, and water-cooling systems. The laser beam was focused at a distance of 13 mm above the substrate surface. Argon was employed as the shielding gas to protect the melt pool from oxidation and to cool the laser head. The processing sequence is illustrated in Figure 1e.

The fixed boundary conditions were the degrees of freedom of the four lines of the cuboid model in the x , y , and z directions. Figure 1f shows the mesh generation result, consisting of ~20,000 finite elements. The laser heat source is located at the top of the repaired layer. The LDED process parameters are listed in Table 2. Nine orthogonal experiments were designed to investigate the effects of powder feed rate, laser power, and scanning speed. The linear energy density (E_l) during LDED can be calculated using Equation 1:²⁵

$$E_l = \frac{p}{v} \quad (1)$$

where p is the laser power (W), and v is the scanning speed (mm/s).

Table 1. Chemical composition of the FeCoCrNiAl powder and matrix (wt.%)

Chemical	Composition							
GCr18Mo	Copper	Manganese	Phosphorus	Chromium	Nickel	Molybdenum	Copper	Iron
	1.0	0.3	0.01	1.7	0.1	0.2	0.1	Bal.
FeCoCrNiAl	Iron	Cobalt	Chromium	Nickel	Aluminum	Oxygen	Nitrogen	Carbon
	21.9	23.2	20.5	23.6	10.8	0.02	0.01	0.01

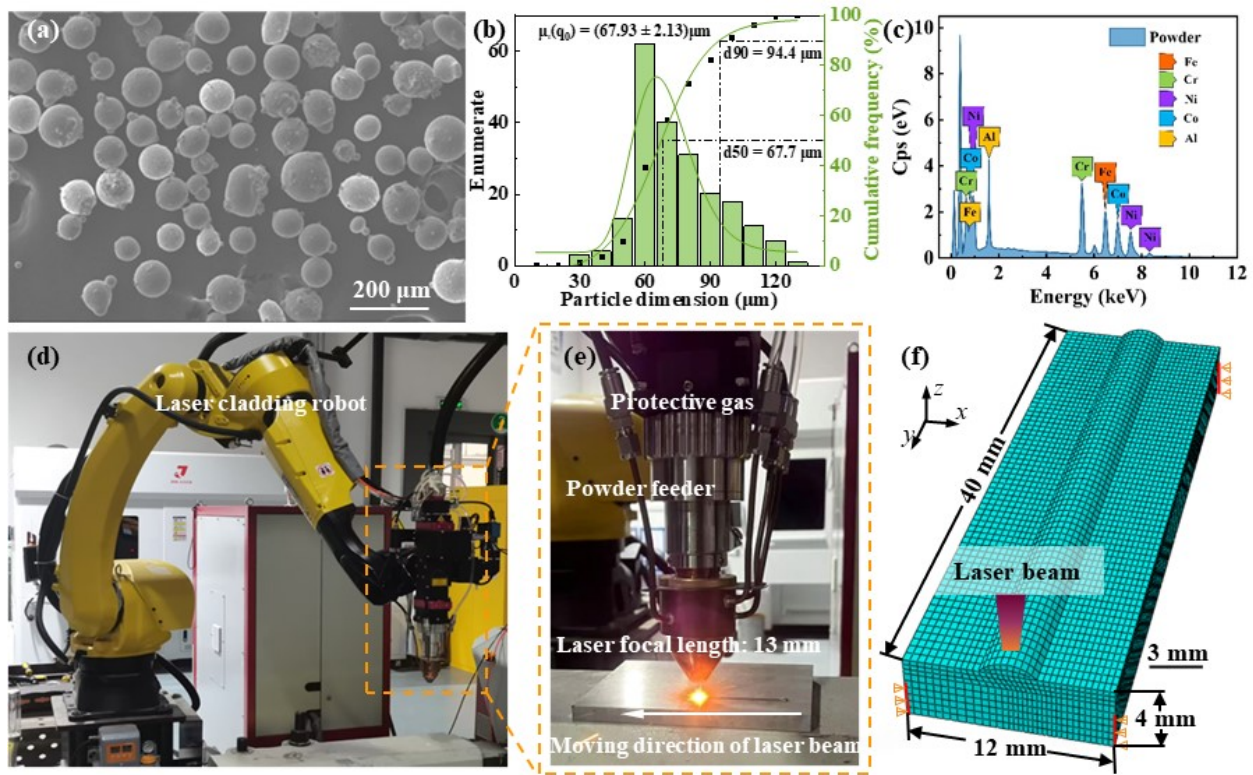


Figure 1. Experimental materials and laser-directed energy deposition (LDED) process. (a) Powder morphology. Scale bar: 200 μm ; magnification: 100 $\times$. (b) Particle size distribution. (c) Powder element content. (d) The LDED systems. (e) Manufacturing process. (f) Simulation mesh model.

Table 2. Laser-directed energy deposition processing experimental design

No.	Powder feed rate (f , g/min)	Scanning speed (v , mm/s)	Laser power (P , W)	Linear energy density (E_p , J/mm)
N1	10.4	5	1,700	340
N2	10.4	6	2,000	333
N3	10.4	7	2,300	328
N4	11.8	5	2,000	400
N5	11.8	6	2,300	383
N6	11.8	7	1,700	242
N7	13.6	5	2,300	460
N8	13.6	6	1,700	283
N9	13.6	7	2,000	285

2.2. Simulation

In this work, a double-ellipsoidal Gaussian heat source model was used to represent laser energy input. The surface heat flux acting on the top surface is expressed as follows (Equation 2):²⁶

$$Q(x, y, z, t) = \frac{6\sqrt{3}P}{abc\pi\sqrt{\pi}} \exp\left(-\frac{3x^2}{b^2} - \frac{3y^2}{c^2} - \frac{3z^2}{a^2}\right) \quad (2)$$

where, P is the laser power (W), a , b , and c are the characteristic heat-source dimensions along the x , y and z directions (m), x , y , and z are the spatial coordinates of the heat-source center in the moving coordinate system, and t accounts for the temporal evolution as the laser moves along the scanning path. The transient temperature field was described by a temperature-dependent heat conduction equation, in which the thermal conductivity and specific heat vary with temperature. The governing equation can be written as Equation 3:²⁷

$$\rho c(T) \frac{\partial T}{\partial t} = \nabla \cdot (k(T) \nabla T) + Q(x, y, z, t) \quad (3)$$

where, ρ is the material density, $c(T)$ is the temperature-dependent specific heat capacity, $k(T)$ is the temperature-dependent thermal conductivity, and $Q(x, y, z, t)$ denotes the internal heat source associated with the moving laser.

The mechanical response of the material was characterized using a temperature-dependent linear elastic model coupled with thermal expansion (Equation 4):²⁶

$$\varepsilon^{th}(T) = \int_{T_0}^T \alpha(\theta) d\theta \quad (4)$$

The elastic stress–strain constitutive relation is given by **Equation 5**:²⁶

$$\sigma = D(T) : (\varepsilon - \varepsilon^{th}(T)) \quad (5)$$

where, $D(T)$ is the temperature-dependent elasticity matrix constructed from Young's modulus, $E(T)$ and Poisson's ratio, $\nu(T)$. Since the plastic data provided only a single yield point without a hardening curve, the material's response in the present simulations is predominantly quasi-elastic over the investigated temperature range.

$$\begin{bmatrix} \sigma_{11} \\ \sigma_{22} \\ \sigma_{33} \end{bmatrix} = D(T) \left(\begin{bmatrix} \varepsilon_{11} \\ \varepsilon_{22} \\ \varepsilon_{33} \end{bmatrix} - \varepsilon^{th}(T) \begin{bmatrix} 1 \\ 1 \\ 1 \end{bmatrix} \right) \quad (6)$$

Equation 6 represents the thermoelastic constitutive relation used to compute the normal stress components σ_{11} , σ_{22} , and σ_{33} . The stress vector was obtained by multiplying the temperature-dependent elasticity matrix $D(T)$ with the difference between the total strain and the thermal strain. The term $\varepsilon^{th}(T)[1,1,1]^T$ denotes the free thermal expansion, which does not contribute to stress unless it is constrained. The equivalent von Mises stress was computed as **Equation 7**:

$$\sigma_v = \sqrt{\frac{1}{2}[(\sigma_{11} - \sigma_{22})^2 + (\sigma_{22} - \sigma_{33})^2 + (\sigma_{33} - \sigma_{11})^2 + 6(\tau_{12}^2 + \tau_{23}^2 + \tau_{31}^2)]} \quad (7)$$

which is used to assess the magnitude and distribution of thermally induced stresses during the heating and cooling cycles. To accurately reproduce the thermal-stress evolution during cladding, the parameters listed in **Table 3** were implemented to model heat conduction and stress development. The material model parameters used in this study are listed in Table S1.

Table 3. Thermophysical parameters in simulation²⁸

Physical property	Value	Units
Absolute zero temperature	−273.15	°C
Stefan–Boltzmann constant	5.67×10^{-8}	W m ^{−2} K ^{−4}
Film coefficient	20	m ^{−2} K ^{−1}
Sink temperature	20	°C
Emissivity	0.85	-
Ambient temperature	20	°C
Initial temperature	20	°C

2.3. Characterization

The distribution of elements and precipitates near the crack was studied using a scanning electron microscope (SEM; Sigma300, Zeiss, Germany) equipped with an energy-dispersive X-ray spectroscopy (EDS) detector. Point EDS analyses were performed at an accelerating voltage of 15 kV to determine the elemental composition of the precipitates and crack regions. The samples were subjected to electrochemical etching prior to SEM observation.

Electron backscatter diffraction (EBSD) was employed to analyze grain distribution, orientation misorientation, and dislocation accumulation in the vicinity of cracks. Grain analysis was performed using AZtecCrystal software (version 2.1). The EBSD acquisition parameters included a step size of 4 μ m and an accelerating voltage of 15 kV. Optical microscopy (OM; VHX-X1F, Keyence, Japan) was used to characterize the cross-sectional microstructure of the deposited coatings. This work used a hardness tester to evaluate the microhardness from the molten pool to the matrix (MHVS-10AT, Beijing Times Guangnan Technology Co., Ltd., China). Tests were performed under a constant load of 4.9 N, with each point held for 5 seconds. Measurements were taken every 100 μ m downwards from the coating surface across the coating cross section. Testing was stopped at three points in the matrix region (<220 HV_{0.5}). The interval between each row of hardness tests was 100 μ m, and six rows were tested per sample.

3. Results

3.1. Surface morphology and internal defects

Surface and internal defects in the single-pass molten pool are key indicators for evaluating the quality of LDED repairs of GCr18Mo bearing steel. In particular, cracks in the molten pool severely limit component serviceability; thus, clarifying the crack characteristics of the LDED-manufactured FeCoCrNiAl HEAs. **Figure 2a** shows that the surface quality of the nine sets of single-pass molten pools fabricated by LDED is excellent, free from discontinuities and lack-of-fusion defects. **Figure 2b–j** show the internal defects of the molten pool under different process parameters. An increase in the linear laser energy density from 328 J·mm^{−1} to 340 J·mm^{−1} triggered a pronounced transition in the internal defects of the molten pool at a $f = 10.4$ g·min^{−1}, from gas pores to severe cracking, primarily comprising solidification cracks and some HAZ cracks. Li *et al.*²⁹ reported laser additive manufacturing of the nickel-based superalloy DZ125L, during which HAZ cracking occurred, and incoherent carbide particles were observed at the crack sites. These observations are consistent with those reported in the present work. Notably, the solidification cracks exhibited

a propagation tendency from the molten pool top to the matrix, as shown in Figure 2d. Furthermore, the HAZ cracks were predominantly distributed along the molten pool boundaries (i.e., heterogeneous interfaces³⁰). Mei *et al.*³¹ also observed a similar phenomenon in the additively manufactured Inconel 718 alloy. The internal integrity of the molten pool underwent a pronounced transformation with the escalation of linear laser energy density at a $f = 11.8 \text{ g} \cdot \text{min}^{-1}$, transitioning sequentially from randomly dispersed pores, to severe HAZ cracking, and ultimately to a nearly defect-free regime. The solidification and HAZ cracks are effectively improved with a line laser energy density of $283 \sim 460 \text{ J} \cdot \text{mm}^{-1}$ ($f = 13.6 \text{ g} \cdot \text{min}^{-1}$). The solidification crack lengths at the top of the molten pool were reduced, while the crack density in the HAZ was decreased. The main defect types of the LDED repairs of GCr18Mo bearing steel are solidification and HAZ cracks.

Low f and suitable E_l matching can effectively eliminate cracks, as shown in Figure 2b and 2g. Solidification cracks account for a significant proportion of defects in the LDED-manufactured FeCoCrNiAl HEAs. Therefore, clarifying the initiation and propagation mechanisms of the cracks is crucial for achieving high-quality repair.

3.2. Stress field evolution

Extrusion caused by thermal expansion deformation can lead to cracks in FeCoCrNiAl HEAs prepared by the LDED process. Therefore, the initiation and propagation mechanisms of solidification and HAZ cracks can be evaluated by analyzing the temperature and stress distribution. Figure S1 verifies the model's effectiveness by comparing the temperature field with that of the actual sample. Figure 3a and 3d show the temperature distribution at $t = 3.9$ seconds and von Mises stress distribution at $t =$

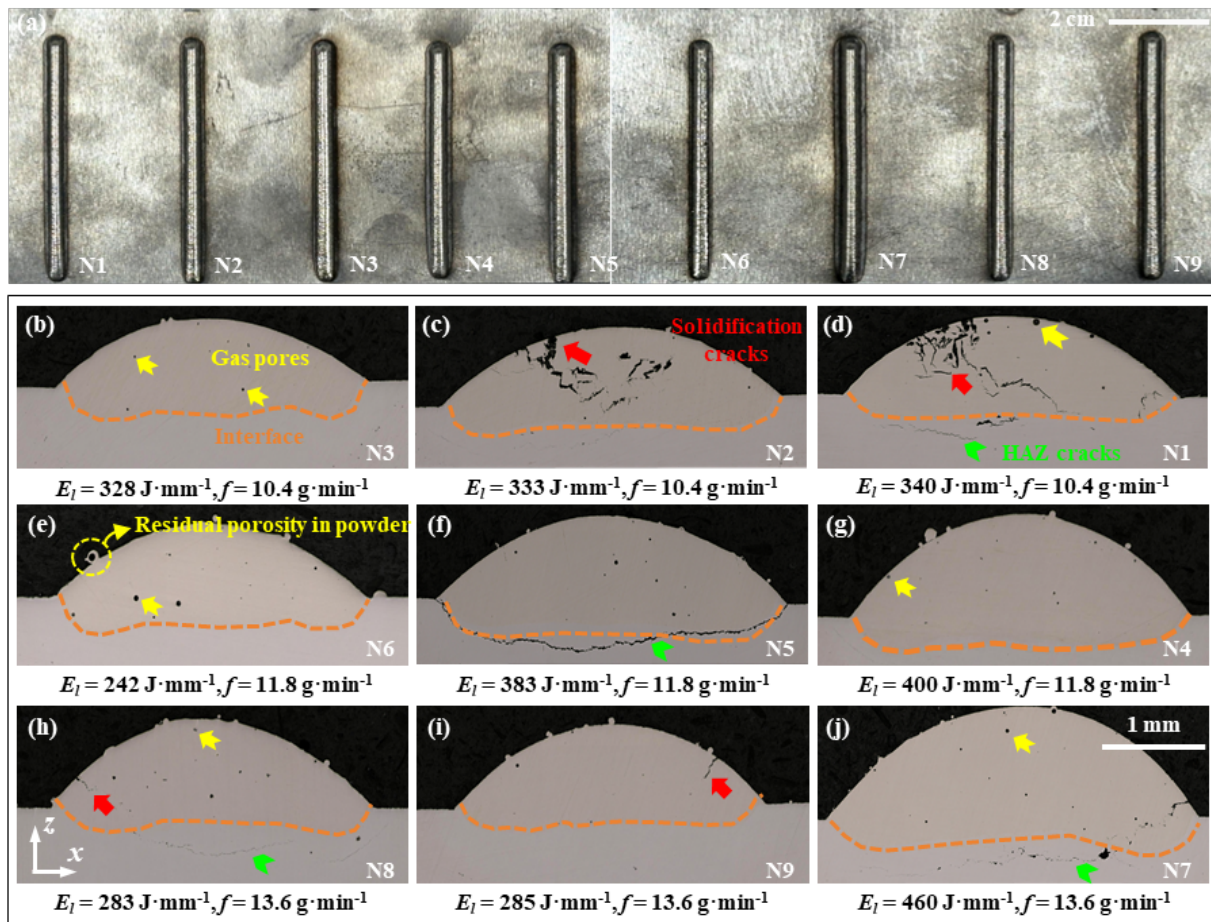


Figure 2. Single-pass molten pool forming quality of the FeCoCrNiAl high-entropy alloy fabricated by laser-directed energy deposition. (a) Surface morphologies. Scale bar: 2 cm. (b–j) Internal defects of the molten pool under different process parameters: (b) $E_l = 328 \text{ J} \cdot \text{mm}^{-1}$, $f = 10.4 \text{ g} \cdot \text{min}^{-1}$, (c) $E_l = 333 \text{ J} \cdot \text{mm}^{-1}$, $f = 10.4 \text{ g} \cdot \text{min}^{-1}$, (d) $E_l = 340 \text{ J} \cdot \text{mm}^{-1}$, $f = 10.4 \text{ g} \cdot \text{min}^{-1}$, (e) $E_l = 242 \text{ J} \cdot \text{mm}^{-1}$, $f = 11.8 \text{ g} \cdot \text{min}^{-1}$, (f) $E_l = 383 \text{ J} \cdot \text{mm}^{-1}$, $f = 11.8 \text{ g} \cdot \text{min}^{-1}$, (g) $E_l = 400 \text{ J} \cdot \text{mm}^{-1}$, $f = 11.8 \text{ g} \cdot \text{min}^{-1}$, (h) $E_l = 283 \text{ J} \cdot \text{mm}^{-1}$, $f = 13.6 \text{ g} \cdot \text{min}^{-1}$, (i) $E_l = 285 \text{ J} \cdot \text{mm}^{-1}$, $f = 13.6 \text{ g} \cdot \text{min}^{-1}$, and (j) $E_l = 460 \text{ J} \cdot \text{mm}^{-1}$, $f = 13.6 \text{ g} \cdot \text{min}^{-1}$. Scale bar: 1 mm; magnification: 100x.

608 seconds under N1 and N5 processes. The difference in E_t between the N1 and N5 processes is $43 \text{ J} \cdot \text{mm}^{-1}$. Figure 3b and e show that the highest temperature of the molten pool in the N1 process is $3,600^\circ\text{C}$, while the highest temperature of the molten pool in the N5 process is $4,200^\circ\text{C}$. During the process, the N5 process exhibited a stress fluctuation lasting ~ 0.4 seconds within the molten pool, characterized by a peak stress amplitude of 36 MPa . In contrast, the N1 process showed a shorter fluctuation duration of about 0.28 seconds, with a stress amplitude of 8 MPa . The N5 process took 0.9 seconds to cool the temperature from $1,850^\circ\text{C}$ to $1,000^\circ\text{C}$, while the N1 process took 0.85 seconds to cool the temperature from $1,650^\circ\text{C}$ to $1,000^\circ\text{C}$. Differences in cooling rates led to differences in the magnitude of stress fluctuations. Figure 3c and 3f show that the stress under N1 fluctuated downward by 547.5 MPa , then recovered over a duration of 1.4 seconds, while under N5, the stress fluctuated downward by 628.8 MPa and recovered in 1.1

seconds. Larger stress fluctuations and faster rate of change are key factors leading to crack formation in the HAZ.³² The von Mises stress distribution in the molten pool and matrix for processes N1 and N5 is shown in Figure 3g and 3h. The von Mises stress in the N1 process coating exceeded that in the N5 process, with a maximum stress difference of 79 MPa between the centers of the two molten pools. Moreover, in the N5 process, the von Mises stress at both ends of the molten pool was higher than that in the central region. This stress distribution pattern closely corresponds with the crack distribution observed in the experiment. Figure 3h shows that the data distribution trends of the N5 and N1 processes were consistent. The von Mises stress on both sides of the molten pool under the N5 process was higher than that under the N1 process, with a difference of 71 MPa . The higher von Mises stress on both sides in the N5 process is in agreement with the observed distribution of HAZ cracks in the actual experiments.

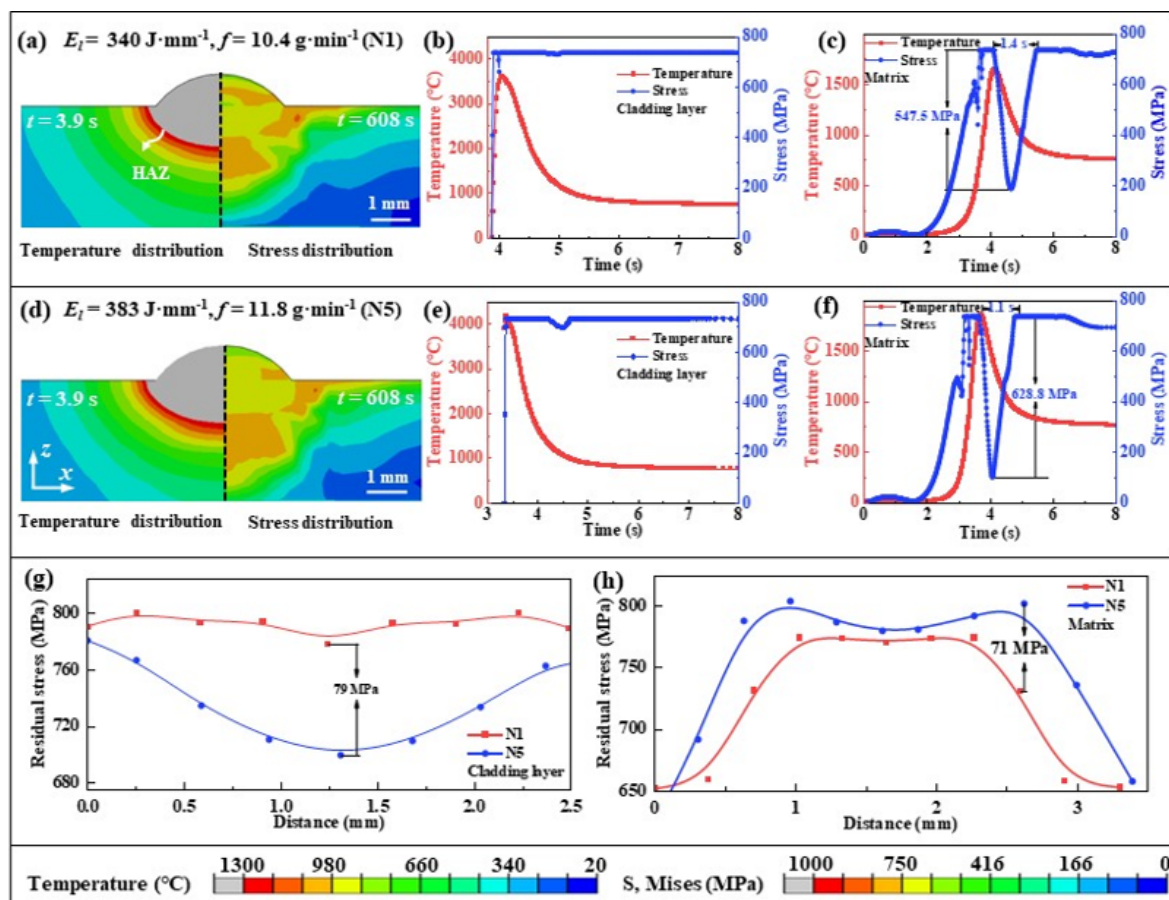


Figure 3. Thermal-mechanical coupled simulation analysis of the molten pool in FeCoCrNiAl high-entropy alloy fabricated by laser-directed energy deposition process. (a) Simulated temperature and stress field of the N1 process. (b) Variations of stress and temperature with time in the N1 molten pool. (c) Variations of stress and temperature with time in the N1 matrix. (d) Simulated temperature and stress field of the N5 process. (e) Variations of stress and temperature with time in the N5 molten pool. (f) Variations of stress and temperature with time in the N5 matrix. (g) Stress comparison of the molten pool between N1 and N5. (h) Stress comparison of the matrices between N1 and N5.

This work clarifies the direction of crack initiation and propagation by obtaining the distribution data of the three principal stresses (S11, S22, S33) in von Mises stress. Figure 4a–c show the simulation analysis results under the N1 process, revealing significant differences in stress distribution between the molten pool and the matrix: the molten pool predominantly experienced compressive stress in S11. Along S22, the ends of the molten pool were subjected to compressive stress while the middle region exhibited tensile stress. In S33, the ends were nearly stress-free, with the central region also showing tensile stress. The S11 and S22 stresses in the matrix near the HAZ both exhibited tensile characteristics. The distribution of S33 stress was consistent with that of the molten pool, with the ends showing compressive stress and the central region experiencing tensile stress. The extraction and analysis of stress data should primarily focus on whether the stress values surpass the material's yield strength. Figure 4d–f show that thermal stress alone is insufficient to induce cracking in the molten pool. The regions of stress concentration were mainly distributed in the center of the molten pool (The yield strength of FeCoCrNiAl HEAs is ~ 450 MPa³³). The principal stresses at the edges of the molten pool were predominantly compressive, which can effectively inhibit crack propagation. Figure 4c and 4d illustrate that solidification cracks originate around the central area of the molten pool, with relatively severe cracking. In contrast, Figure 4h and 4i show that while cracks do occur at the edges of the molten pool, the three principal stresses in these regions were all compressive, effectively suppressing crack initiation. Consequently, the molten pool in these areas exhibited significant improvement. Figure 4j–l show that the stress in the HAZ of the matrix is greater than that in the molten pool. The S22 stress in the central region exceeded 850 MPa (The yield strength of GCr18Mo steel at room temperature exceeds 850 MPa³⁴). The S11 stress at the edge of the matrix was 420 MPa, and the S11 stress at the center of the matrix was 714 MPa. This stress can lead to crack initiation at the center of the matrix. The S33 stress in the matrix was -150 MPa, and the S33 stress in the central region was 270 MPa. Because the S33 stress is compressive stress at the edge of the matrix, it has an inhibitory effect on crack initiation. The crack growth orientation in the HAZ on the xz plane exhibited an overall horizontal distribution and preferentially appeared at the center of the matrix. Figure 4d, 4f, and 4h show the HAZ crack morphology, which is consistent with the above findings.

3.3. Crack feature

Figure 5a shows the microstructure of the LDED-manufactured FeCoCrNiAl HEAs. Figure 5b shows the

grain orientation distribution around multiple cracks. These cracks were all short and straight, approximately $130\text{ }\mu\text{m}$ in length. Figure 5c shows a $378\text{ }\mu\text{m}$ long crack with two bends and the surrounding grain orientation distribution. Two cracks (L1 and L2) were selected in zone 1, and one crack (L3) was selected in zone 2. The grain orientation difference around each crack and the grain orientation difference at the start and end of the broken line segment were statistically analyzed, as shown in Figure 5d. The grain orientation difference on both sides of cracks L1 and L2 was small ($<2^\circ$), indicating that the crack in zone 1 is a trans-granular crack. For crack L1, the local grain orientation difference measured downward from the top of the molten pool was 19° . As the crack propagated toward the mid region, the surrounding grains exhibited orientation differences of less than 10° . Crack propagation along grain boundaries is driven by a reduction in energy driving force, and it ultimately ceases when the orientation differences on opposing sides reach 25° and 46° , respectively.³⁵ For crack L2, the grain orientation difference measured from left to right increased from about 2° to 9° , and then stopped at 45° . For crack L3, the grain orientations around it from top to bottom were all greater than 15° and showed a gradual increasing trend, which is a typical characteristic of inter-granular cracks. Figure 5e shows the overall grain boundary distribution characteristics of the molten pool. The central region of the molten pool is dominated by high-angle grain boundaries, while the edges exhibit a large number of low-angle grain boundaries. The high-angle grain boundaries accounted for 64.9%, and low-angle grain boundaries accounted for 35.1%. Interestingly, the low-angle grain boundaries appearing near the cracks in the central part of the molten pool were more likely related to dislocation structures and sub-grain structure evolution generated during solidification.³⁶ Figure 5f–h show the Schmid factor (SF) distribution of the molten pool. Figure 5g shows that the SF in the upper region of crack L1 was 0.49, and the value at the end of the L1 crack was 0.41. Figure 5h shows that the inter-granular crack completely avoided all regions with relatively low dislocation density.³⁷ The L3 crack initiated from the upper left corner, passed through the middle region with SF values between 0.33 and 0.30, and finally avoided the region with SF of 0.39, propagating downward and rightward until arrest. The grains around the crack were prone to plastic deformation, which promoted crack initiation and propagation.³⁸

Kernel average misorientation (KAM) generally serves as an indicator of local lattice curvature and is defined as the average misorientation between a kernel point and its neighboring points, excluding those beyond the grain boundary. Generally, higher KAM values indicate severe deformation, which is associated with an increased

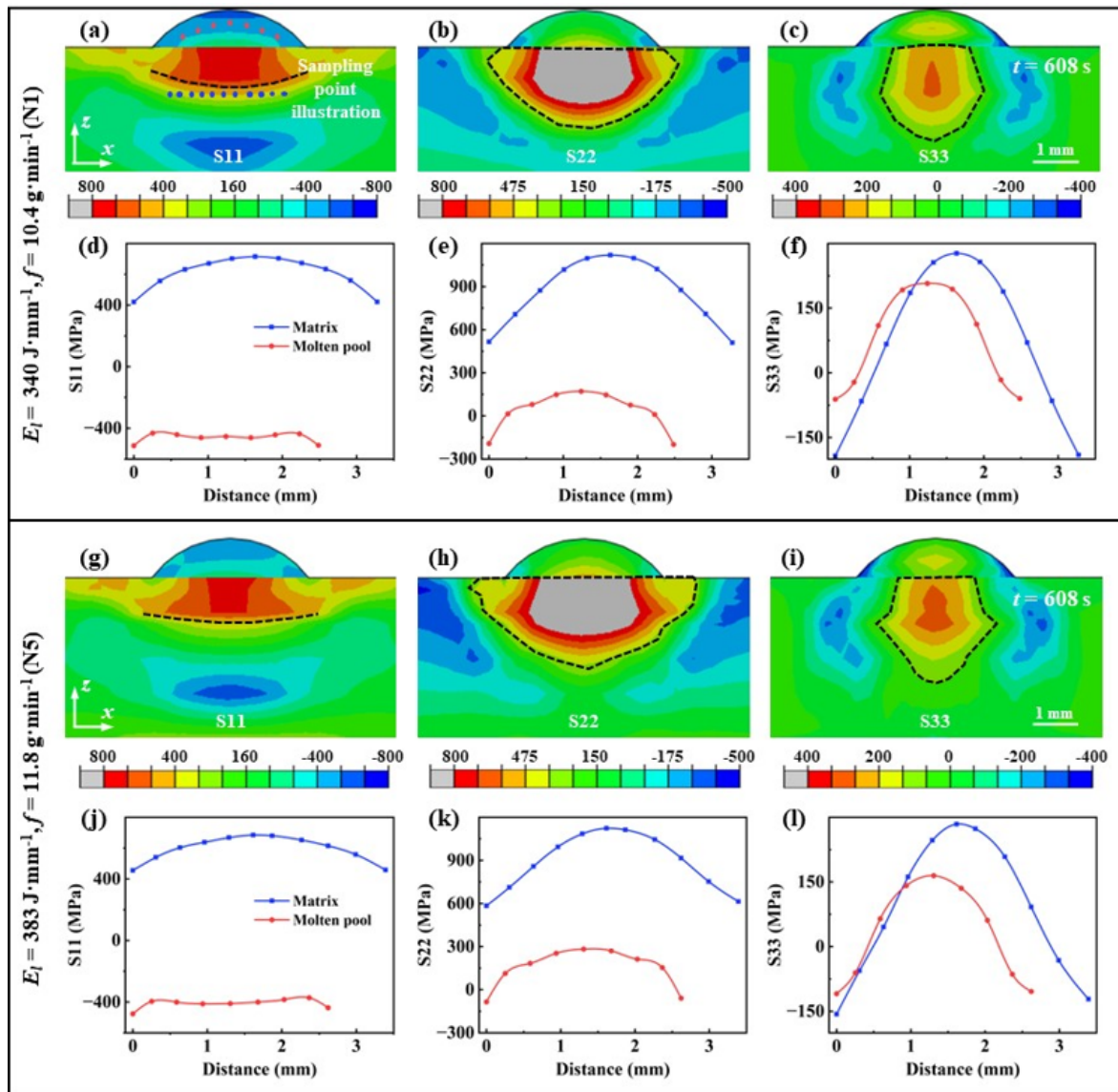


Figure 4. The principal stress in the molten pool of the laser-directed energy deposition-manufactured FeCoCrNiAl high-entropy alloys. (a) S11, (b) S22, (c) S33 residual stresses field contour map, (d–f) S11, S22, and S33 residual stresses evolution with distance ([d] S11, [e] S22, [f] S33 stress, N1 process), (g) S11, (h) S22, (i) S33 residual stresses field contour map, and (j–l) S11, S22, and S33 residual stresses evolution with distance ([j] S11, [k] S22, [l] S33 stresses, N5 process).

dislocation density.³⁹ Figure 6a presents the local lattice deformation characterized by EBSD within the molten pool. A high KAM was concentrated around the cracks in the molten pool, suggesting strong local orientation gradients induced by crack-related plastic deformation. As this work focuses on the stress effects associated with crack formation, it should be noted that KAM alone cannot quantitatively represent residual stress. Therefore, the density of the geometrically necessary dislocations (GNDs) has a physical relationship with strain gradients and stress concentrations, and can be used to explain crack-related

stress fields (Equation 8):⁴⁰

$$\rho_{\text{GND}} = \frac{\alpha \theta}{bx} \quad (8)$$

where θ is the threshold orientation deviation angle, x is the acquisition step size, b represents the magnitude of the Burgers vector, and α is a material-related constant, typically ranging from 2 to 5.1.^{41,42} To assess the impact of parameter selection on the order of magnitude, we conducted a sensitivity analysis for $\alpha = 2\text{--}5.1$; α only affects the absolute scaling of GND, without changing

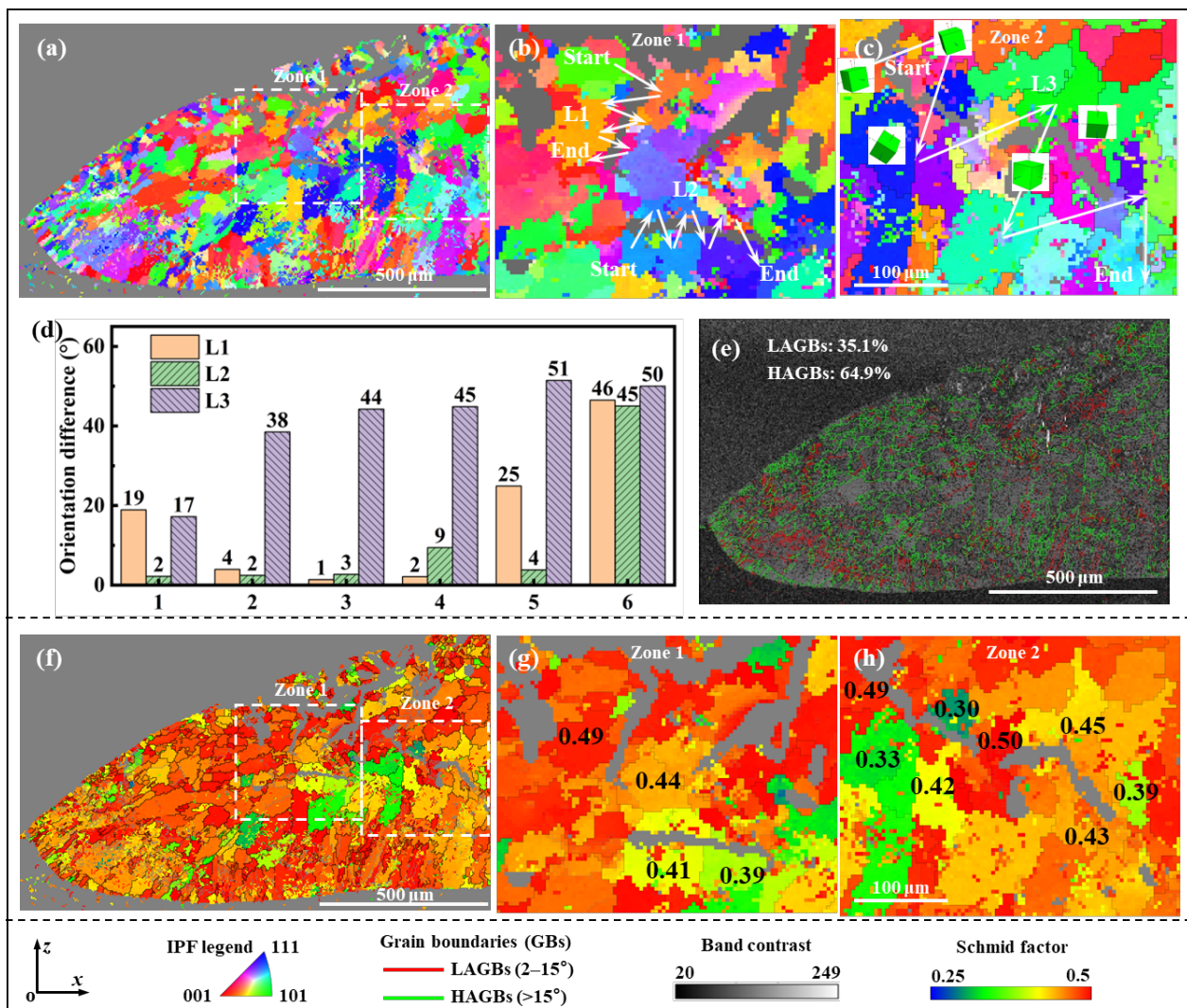


Figure 5. Electron backscatter diffraction maps in the xz plane of the laser-directed energy deposition-manufactured FeCoCrNiAl high-entropy alloys. (a–c) Inverse pole figure (IPF) maps: (a) overall zone (scale bar: 500 μm), (b) zone 1, (c) zone 2 (scale bar: 100 μm); (d) line measurement orientation difference distribution; (e) the map of grain boundaries and band contrast (scale bar: 500 μm ; magnification: 40 $\times$); and (f–h) Schmid factor maps: (f) overall zone (scale bar: 500 μm), (g) zone 1, and (h) zone 2 (scale bar: 100 μm).

the spatial distribution or the comparison conclusions. In this work, a sensitivity analysis was performed, and $\alpha = 3$ was selected as it provides a balanced and physically reasonable magnitude of GND density while maintaining a clear spatial distribution consistent with EBSD–KAM maps.⁴³ Figure 6c shows the GND density values around the crack in zone 1. Line measurements were performed on cracks L1 and L2 in the figure, with a line width of 15 pixels. Figure 6d shows the GND density data for crack L1. The average GND density in this region was $0.51 \times 10^{14} \text{ m}^{-2}$. The GND density distribution near crack L1 showed a trend of high at both ends and low in the middle. The intermediate

value of $0.84 \times 10^{14} \text{ m}^{-2}$ was due to stress concentration in this region. The highest GND density for crack L1 was located above the crack, with a value of $0.87 \times 10^{14} \text{ m}^{-2}$. Figure 6e shows the GND density data for crack L2. When measuring the GND density data of crack L2, the data from both ends of crack L2 are statistically analyzed together and shown in Figure 6e. The results indicate that the GND density distribution around crack L2 exhibits a pattern similar to that observed around crack L1. The crack exhibited a GND density distribution pattern that is high at both ends and low in the middle. The highest GND density was $1.33 \times 10^{14} \text{ m}^{-2}$ at the left end and $1.11 \times 10^{14} \text{ m}^{-2}$ at the right

end. The overall GND density was $0.63 \times 10^{14} \text{ m}^{-2}$. The crack morphology in Figure 6f differs from that of cracks L1 and L2 in Figure 6c. The crack in Figure 6f shows two bends. The overall crack was divided into three segments. The GND density of the matrix was $0.32 \times 10^{14} \text{ m}^{-2}$. Linear measurements were performed on each segment, with a width of 15 pixels. The

average GND densities of these three segments were $0.54 \times 10^{14} \text{ m}^{-2}$, $0.69 \times 10^{14} \text{ m}^{-2}$, and $0.63 \times 10^{14} \text{ m}^{-2}$, respectively. Combining these three segments reveals that the GND density was higher at their junctions, with the maximum value ($1.12 \times 10^{14} \text{ m}^{-2}$) occurring at the junction of Line 1 and Line 2. The upward fluctuation in GND density value at the center of the

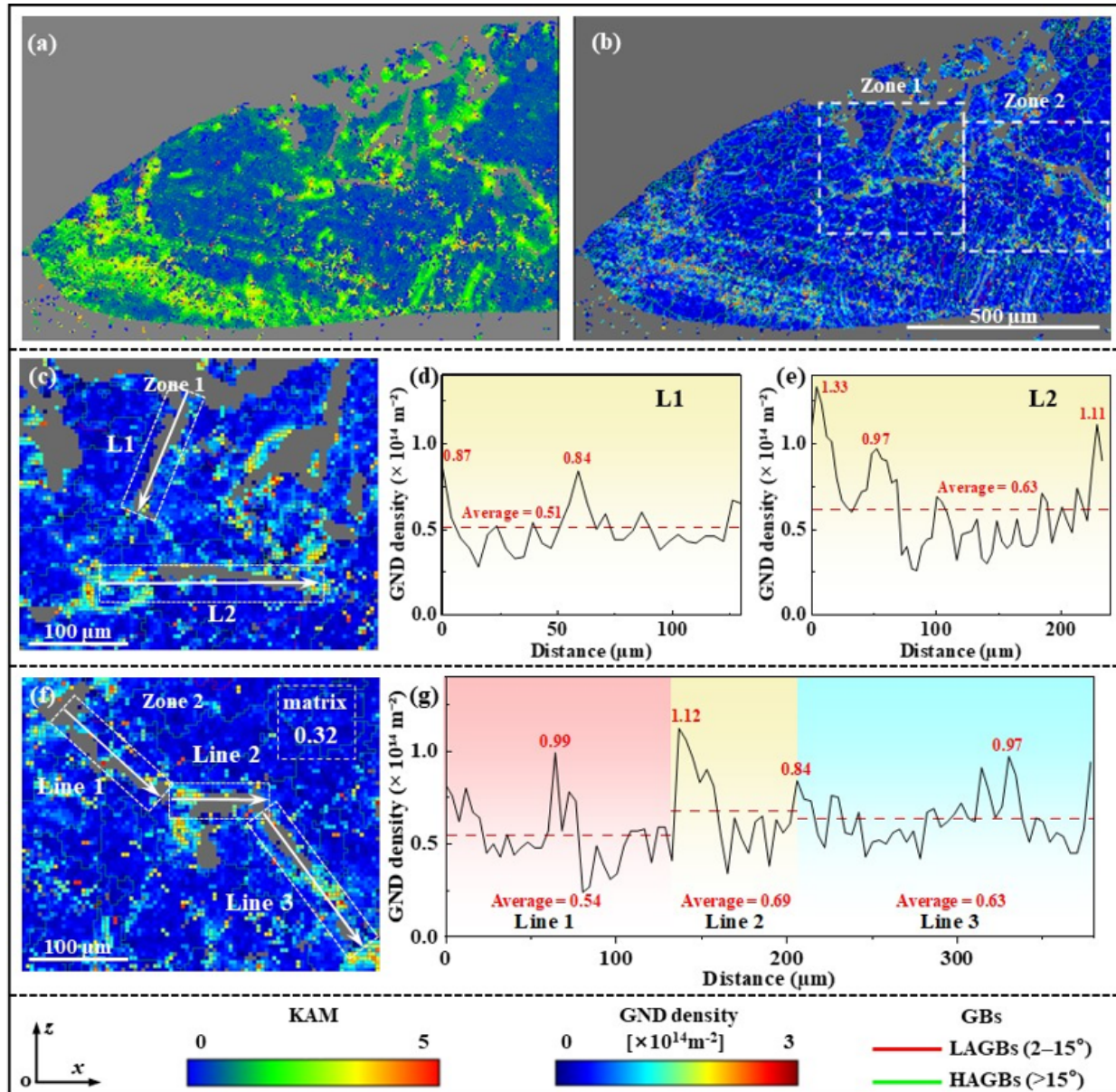


Figure 6. Stress and dislocation density of the laser-directed energy deposition-manufactured FeCoCrNiAl high-entropy alloys. (a) Kernel average misorientation (KAM) map. Scale bar: 500 μm; magnification: 40×. (b) The geometrically necessary dislocation and grain boundary map. Scale bar: 500 μm; magnification: 40×. (c) Geometrically necessary dislocation map of zone 1. Scale bar: 100 μm. (d) Geometrically necessary dislocation value measured by L1. (e) Geometrically necessary dislocation value measured by L2. (f) Geometrically necessary dislocation map of zone 2. Scale bar: 100 μm. (g) Geometrically necessary dislocation value near the crack.

crack was due to stress concentration in that region.

Figure 7 shows the cracks and surrounding element distribution characteristics for processes N1 and N5, respectively. Figure 7a shows an SEM image of solidification cracks. Solidification cracks exhibited a distribution characteristic of extending from the upper left of the molten pool toward the center of the molten pool. Figure 7b shows the element distribution from the top of the molten pool down to the matrix. The three cracks selected by the line scan second within the molten pool region corresponded to three regions exhibiting elemental anomalies. The weak carbon element observed in the first and third cracks was consistent with carbon exposure occurring after crack formation, implying that precipitated carbon was unlikely to serve as a primary stress source for microstructural displacement in these regions. By contrast, the distinct carbon spike detected at the intersection of solidification cracks at the second anomalous point reflects localized carbon enrichment, rather than a homogeneous carbon distribution throughout the molten pool. The transition region between the molten pool and the matrix was approximately 77 μm . As shown in Figure 7c, elemental mapping was performed in zone 1. The results indicate a clear enrichment of carbon element along the crack regions. Furthermore, point EDS analyses were conducted at positions P1–P4, where the measured carbon element content exceeded 27 wt.%. Figure 7d shows the SEM image of the HAZ crack, located at the interface. The signal of FeCoCrNiAl HEAs was relatively weakened, and it mainly transformed into the signal of GCr18Mo steel with iron as the core element. According to previous studies,^{44,45} the aluminum element enrichment in FeCoCrNiAl alloys promotes the phase transformation from face-centered cubic (FCC) to BCC/B2 structures. Niu *et al.*⁴⁶ demonstrated that aluminum-containing FeCoNiCrAl HEAs exhibit a composition-dependent phase transition, where higher aluminum content stabilizes a BCC/B2 phase, while lower aluminum content promotes FCC formation. Therefore, local aluminum segregation can lead to the formation of BCC/B2 phases in the segregated regions. It is also worth noting that the rapid solidification and steep thermal gradients inherent to the LDED process can further intensify elemental segregation, thereby promoting the formation of such phases. These BCC/B2 phases are generally considered to be relatively brittle compared to the FCC matrix. Their presence can deteriorate local mechanical compatibility and act as preferential sites for crack initiation. Based on the EDS results in this work, carbon enrichment

was widely observed along most crack propagation paths, indicating a strong correlation between carbon distribution and crack evolution. However, carbon enrichment was not detected in some crack regions, suggesting that carbon segregation is not universally present in all cases. Therefore, carbon enrichment should not be considered the sole driving force for crack initiation, but rather an important contributing factor to crack formation and propagation. The combined influence of aluminum-induced brittle phases and carbon element enrichment enhances local stress concentration, thereby facilitating crack initiation and propagation.

3.4. Hardness

Liu *et al.*⁴⁷ used a matrix hardness point method to more intuitively evaluate the trend of molten pool hardness variation. Hardness testing was performed at the top center of the molten pool, with six horizontal points spaced 0.1 mm apart. The distribution and data of the hardness test points are shown in Figure 8. The microhardness showed an increasing and then decreasing trend. The average hardness of the matrix was 207.3 $\text{HV}_{0.5}$. The average hardness of the N1, N5, and N9 samples in the molten pool showed a gradually increasing trend, respectively, reaching 442.9 $\text{HV}_{0.5}$, 485.3 $\text{HV}_{0.5}$, and 506.4 $\text{HV}_{0.5}$. The HAZ had an average hardness of 731.2 $\text{HV}_{0.5}$, 683.6 $\text{HV}_{0.5}$, and 712.5 $\text{HV}_{0.5}$. Figure 8a shows the decrease in hardness of the microstructure surrounding the solidification crack. The decreasing hardness trend was positively correlated with the degree of cracking in the surrounding structure. The cracking was most severe in the upper left corner of the crack, and this area had the lowest hardness. The hardness of the area with the most severe cracks was 61.8% lower than that of the surrounding matrix. Figure 8b shows that when the crack appears in the interface area, the hardness error bar of the surrounding hardness is large, especially near the HAZ crack, where the average hardness drops to 672.6 $\text{HV}_{0.5}$. The average hardness above and below this position is 801.3 $\text{HV}_{0.5}$ and 797.3 $\text{HV}_{0.5}$, respectively. Figure 8c shows the hardness distribution cloud map from the N9 process melt pool to the matrix. The overall hardness distribution was uniform, and there were no sudden drops in hardness in certain areas due to defects. In this work, hardness measurements were performed using the Vickers indentation method with a relatively high applied load of 4.9 N. Under such loading conditions, the measured hardness is not governed solely by local dislocation strengthening effects, but is also significantly influenced by the presence of defects such

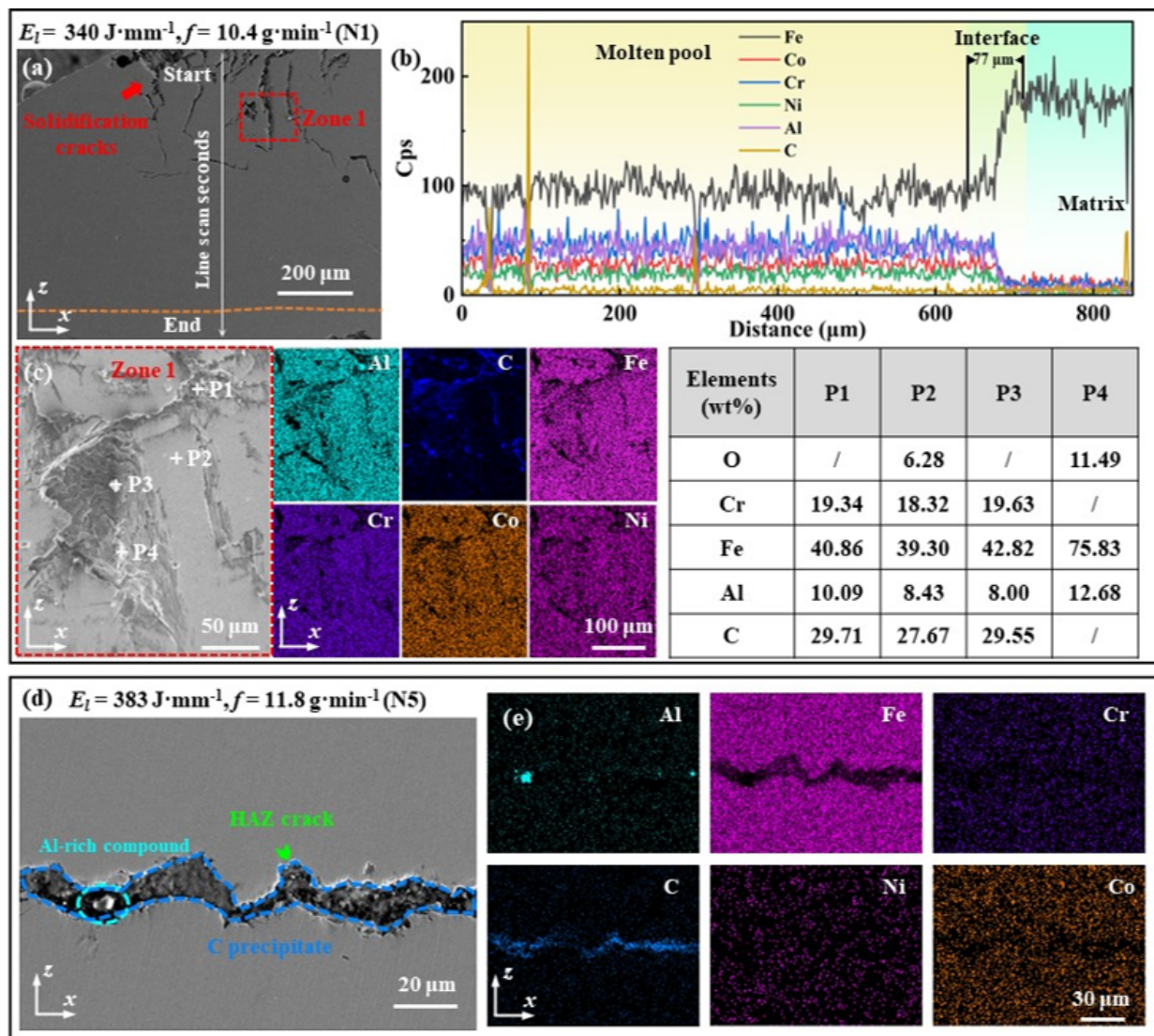


Figure 7. SEM morphologies of cracks in the xz plane and corresponding EDS results of the LDED-manufactured FeCoCrNiAl HEAs. (a–c) N1 process (solidification cracks): (a) SEM image (scale bar: 200 μm ; magnification: 100 $\times$), (b) EDS line scan element distribution results, (c) EDS results (scale bars: 50 μm , 100 μm ; magnifications: 500 $\times$). (d–e) N5 process (HAZ crack): (d) SEM image (scale bar: 20 μm ; magnification: 800 $\times$), (e) EDS results (scale bar: 30 μm ; magnification: 800 $\times$).

Abbreviations: EDS: Energy-dispersive X-ray spectroscopy; HAZ: Heat-affected zone; HEAs: High-entropy alloys; LDED: Laser-directed energy deposition; SEM: Scanning electron microscopy.

as cracks and porosity within the indentation zone. In particular, although increased GND density at the crack tip may contribute to local strain hardening, this effect is counterbalanced and even dominated by the severe reduction in effective load-bearing area caused by crack-induced geometric discontinuities and nearby porosity. These defects lead to stress concentration and premature local failure during indentation, resulting in an apparent reduction in measured hardness.

4. Discussion

4.1. Cracking mechanism of the laser-directed energy deposition-manufactured high-entropy alloys

Random cracks were observed in HEA coatings prepared using the LDED process. These cracks hinder the widespread adoption of the LDED process. Therefore, it is necessary to conduct an in-depth

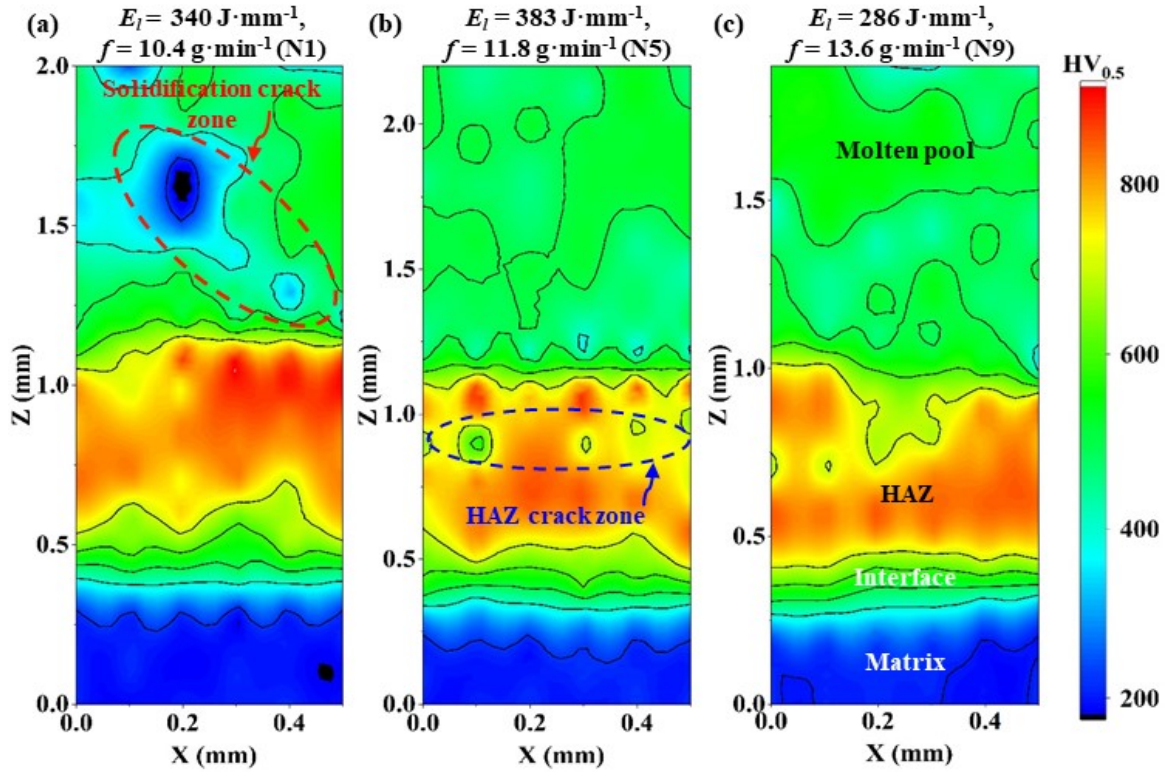


Figure 8. Hardness of the laser-directed energy deposition-manufactured FeCoCrNiAl high-entropy alloy. (a) N1, (b) N5, and (c) N9 samples.

analysis and discussion of the factors that lead to crack initiation, propagation, and cracking during the LDED preparation of the molten pool. Figure 9a shows the location of cracks during the LDED process. The top region of the molten pool experienced the highest cooling rate and the largest temperature gradient, leading to the most severe thermal contraction and tensile stress. As a result, crack initiation is more likely to occur in this region. In addition, during solidification, solute elements tend to segregate into the final solidification zone. Carbon element enrichment was observed in the crack regions near the top of the molten pool (Figure 7c), which weakens the local microstructure and promotes crack formation. Large SF indicates that the resolved shear stress is high and the slip system can be easily activated.⁴⁸ This study revealed a phenomenon related to cracks and SF values: the SF value was higher around the crack. The crack tended to extend and deflect toward areas with higher SF values, as shown in Figure 9b. Ma *et al.*³⁸ also observed and studied the microstructure of crack paths, and found that cracks tend to extend toward grains with smaller twist angles and larger SF. The main reason for the formation of GNDs in the coating prepared by the LDED process is the inconsistent grain

growth rate caused by different temperature gradients in different regions. This leads to compression between grains, resulting in GNDs. Equations 9 and 10 describe the solidification and growth behavior of dendrites:⁴⁹

$$\frac{\partial \phi}{\partial t} = M_\phi \bar{\varepsilon} \left[\nabla \cdot (\varepsilon^2 \nabla \phi) - \frac{\partial}{\partial x} \left(\varepsilon \varepsilon \frac{\partial \phi}{\partial y} \right) + \frac{\partial}{\partial y} \left(\varepsilon \varepsilon \frac{\partial \phi}{\partial x} \right) \right] - M_\phi S + \eta \quad (9)$$

$$\frac{\partial c}{\partial t} + u \cdot \nabla c = \nabla \cdot \left\{ D \left[\nabla c + c(1-c) \times (S_B - S_A) \nabla \phi \right] + j_a \right\} \quad (10)$$

where M_ϕ is the interfacial mobility, S is the normalized entropy, c is the concentration indicator (ranging from 0 to 1), η is the noise term for the generation of random nucleation points, D is the solute diffusivity, and j_a is the antitrapping current introduced by Karma. The expression for S is shown in Equation 11:⁵⁰

$$S_i = W_i g_{(\phi)} + P_{(\phi)} \Delta H_i \left(\frac{1}{T} - \frac{1}{T_m} \right) \quad (11)$$

Birosca *et al.*⁵¹ found that GND depends on the orientation of the surrounding grains. Macroscopic stress and the accumulation of GND are positively

correlated. Dislocations are generated toward the grain boundaries, and high-intensity GND will diffuse to the surrounding area. Figure 9c shows that the different temperature gradients within the molten pool led to inconsistent grain growth rates and orientations, resulting in inconsistent grain orientation differences. The GND value is influenced by both the different grain orientations and the thermal expansion after solidification. GND indicates that this portion of elastic energy is stored. Dislocation generation causes this elastic energy to be released into the surrounding environment.⁵²

Heat-affected zone cracks have been extensively studied; this work focuses on the growth mechanism of solidification cracks. Figure 9d and 9e show the solidification crack initiation and growth process. Holzapfel *et al.*⁵³ found that carbide precipitates strongly influence crack growth by initiating geometrically necessary steps in the crack plane. The solidification crack tends to propagate into grains that are favorably oriented for slip (high SF). A high GND density around the crack and adjacent boundaries indicates intense local strain gradients and strong dislocation storage, reflecting pronounced plastic incompatibility and stress concentration that promote crack deflection

and the development of intragranular damage.^{54,55} The crack evolves from inter-granular crack to trans-granular crack within grains, and the overall crack growth path is biased toward regions where both the SF and GND density are high, corresponding to easy slip orientations coupled with severe local plastic deformation. Finally, the crack stopped growing, forming the solidification crack morphology shown in Figure 9f. The solidification crack exhibited both inter-granular and trans-granular characteristics, as shown in Figure 5. Yang *et al.*⁵⁶ discovered mixed inter-granular/trans-granular fracture in Ti6Al4V alloy during *in situ* tensile testing of the deformation behavior and strengthening mechanism of Ti6Al4V alloy prepared by LDED with iron and trace amounts of boron.

4.2. Relationship between cracks and hardness

In HEA materials, hardness is a crucial indicator characterizing their local resistance to plastic deformation and is closely related to strength. During LDED processing, due to complex thermal and inhomogeneous microstructure, defects such as cracks and porosity are easily generated. These defects weaken the load-bearing capacity and significantly affect the local hardness distribution. Therefore, studying the impact of defects on the material's hardness and

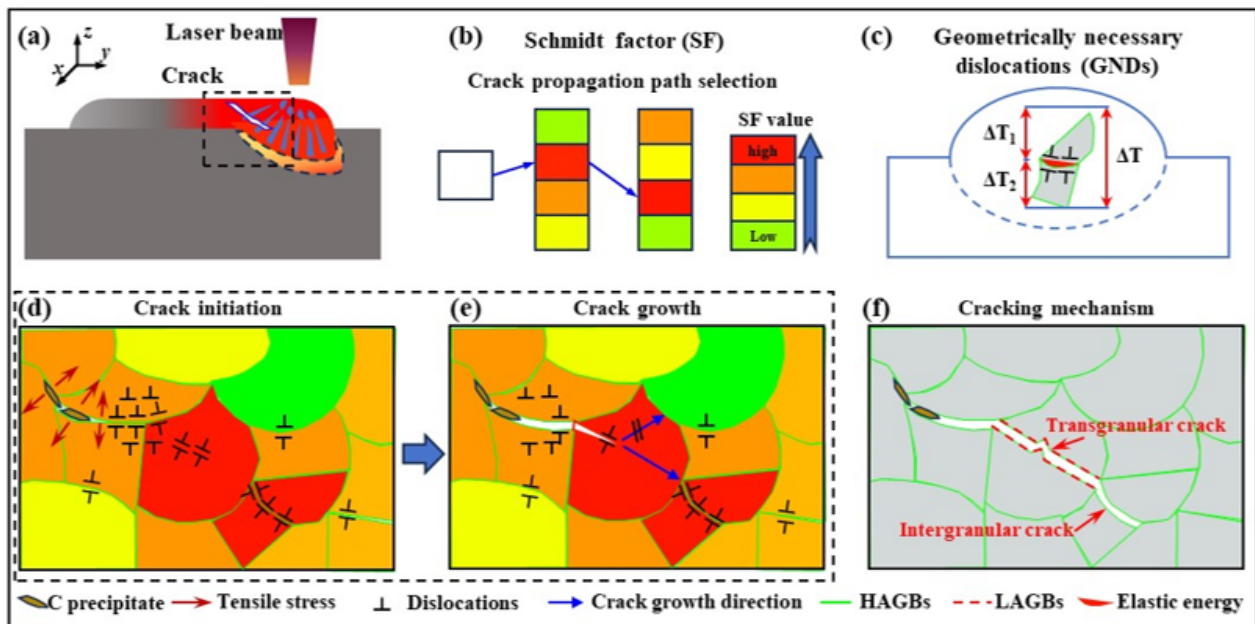


Figure 9. The solidification crack formation mechanism of the laser-directed energy deposition-manufactured FeCoCrNiAl high-entropy alloy coating. (a) Location of cracks during actual processing. (b) Crack growth path selection under the influence of the Schmidt factor. (c) GNDs generation principle. (d) Crack initiation. (e) Crack growth. (f) Cracking mechanism.

mechanical properties is of great significance for evaluating the service performance of repair materials. Kim *et al.*⁵⁷ found that the defects in the material repaired by the DED process resulted in a decrease in yield strength and elongation compared to the forged material. Bi *et al.*⁵⁸ found that due to the special nature of the welding process, the mechanical properties of the welded joint material, especially the uneven yield strength, contain a large number of microcracks. This indicates that there is a linear relationship between the distribution of cracks and the magnitude of the yield strength. From a fracture mechanics perspective, the influence of yield strength on crack tip behavior can be quantitatively described by the plastic zone size ahead of a crack tip, expressed as **Equation 12**:⁵⁸

$$r_p = \frac{1}{\pi} \left(\frac{K_I}{\sigma_{ys}} \right)^2 \quad (12)$$

where, r_p is the crack tip plastic zone size, K_I is the stress intensity factor, and σ_{ys} is the yield strength. This relationship indicates that a reduction in yield strength caused by defects such as cracks leads to an expansion of the plastic zone, thereby promoting localized plastic deformation and premature failure.

Defects such as cracks within an organization can reduce its strength, and hardness is positively correlated with organizational strength. Pavlina *et al.*⁵⁹ statistically analyzed the relationship between yield strength and hardness of various metals, deriving **Equation 13**:

$$H_v = \frac{90.7 + YS}{2.876} \quad (13)$$

where yield strength has units of MPa, and H_v is diamond pyramid hardness, which uses traditional units (kg_f/mm^2). **Figure 8a** and **8b** show that the hardness around the crack decreased. Based on the above findings, we can suggest that cracks affected the yield strength, thus affecting hardness.

5. Conclusion

This work systematically studied the cracking mechanism of the LDED-manufactured FeCoCrNiAl HEA coatings. Numerical simulations were conducted to elucidate the evolution of residual equivalent, tensile, and compressive stresses, establishing the relationship between stress distributions and crack defects within the molten pool and the HAZ zone. Meanwhile, a detailed analysis was conducted on the microstructure at the crack location, including

grain orientation, grain boundaries, precipitates, and dislocations, clarifying the crack initiation and propagation mechanisms. Furthermore, the influence of cracks on hardness was discussed.

- (i) The main defect types in the LDED-manufactured FeCoCrNiAl HEA coatings are solidification and HAZ cracks, with solidification cracks accounting for a significant proportion of defects. Low powder delivery rate (f) and appropriate linear energy density (E_l) matching can eliminate cracks.
- (ii) In the process of preparing HEA coatings using the LDED process, different E_l and f values will lead to different stress evolutions. As the values of E_l and f increase, the stress at the molten pool decreases, while the stress in the HAZ increases, and the crack type in the material changes from solidification crack to HAZ crack.
- (iii) Cracking of the LDED-manufactured FeCoCrNiAl HEA coatings is attributed to the synergistic effects of local grain orientation mismatch, dislocation accumulation, and carbide precipitates. The maximum GND density at the crack tip region ($\sim 1.33 \times 10^{14} \text{ m}^{-2}$) is higher than that of the matrix; high dislocation density provides a strong driving force for crack initiation and growth. Many carbides were present along solidification cracks, whereas aluminum-rich precipitates together with carbides were observed at cracks in the HAZ; the hard carbides promote crack initiation. Solidification cracks within the molten pool exhibit mixed trans-granular and inter-granular fracture mode.
- (iv) The coatings and HAZ exhibited higher overall hardness than the matrix, but cracks led to a decrease in local hardness. Cracks reduced the yield strength of the material, and because yield strength is positively correlated with hardness, they also led to a decrease in local hardness.

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Conflict of interest

The authors declare that they have no competing interests.

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Writing–review & editing: Guang Zeng, Wenjie Liu, Ke Sun

Ethics approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

Availability of data

The data that support the findings of this study are presented in the paper. They are also available from the corresponding authors upon reasonable request.

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