Real-Time Observation of Solidification Microstructure in Laser Remelting Molten Pool: Postprint

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Abstract

Using transparent model alloy succinonitrile-2.0% (mass fraction) ethanol (SCN-2.0%Eth), real-time observations were conducted on the evolution of dendrite growth within the molten pool during laser remelting on a single-crystal substrate (001) plane, with the scanning direction deviating from the substrate [100] crystal orientation at various angles. The study revealed that when scanning parallel to the [100] crystal orientation, the molten pool eventually evolved into parallel symmetric growth of [010] and [01 0] dendrite arrays; when the angle between the scanning direction and the [100] crystal orientation was 20°, the entire molten pool exhibited an asymmetric oscillatory growth mode, wherein one side consistently grew as [01 0] dendrite arrays while the other side displayed competitive alternating growth between [100] and [010] dendrite arrays; when the angle between the scanning direction and the [100] crystal orientation reached 45°, the molten pool consistently exhibited perpendicular growth of [100] and [01 0] dendrite arrays. Based on the dendrite preferred growth criterion, a model describing dendrite growth behavior within the molten pool was established, which well explained the experimental results. The results indicate that the solidification microstructure within the molten pool is jointly influenced by both the molten pool morphology and the substrate crystal orientation.

Full Text

Preamble

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Real-Time Observation of Solidification Microstructure in Laser Remelting Pool

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Abstract

The final quality of parts fabricated by high-energy beam (laser, electron beam, and arc) processing technology is determined by solidification microstructure formation in the molten pool, which has attracted considerable research attention. However, real-time observation of solidification microstructure formation in molten metal pools is extremely difficult due to high temperature, rapid solidification, and opacity. In this work, using a transparent model alloy of succinonitrile-2.0% (mass fraction) ethanol (SCN-2.0%Eth), we conducted real-time observation of solidification microstructure evolution in the molten pool during laser surface remelting on the (001) crystal plane of a single-crystal substrate, with the laser scanning direction deviating at different angles from the [100] crystal orientation of the substrate. It was found that when the scanning direction is parallel to the [100] crystal direction, dendritic columns grow symmetrically in the molten pool, with [010] and [01 0] dendritic columns growing parallel to each other. When the scanning direction deviates by 20° from the [100] crystal orientation, the entire molten pool exhibits an asymmetric oscillatory growth pattern: one side consistently shows [01 0] dendritic column growth, while the other side shows alternating competitive growth between [100] and [010] dendritic columns. When the scanning direction deviates by 45° from the [100] crystal orientation, the molten pool consistently exhibits perpendicular growth between [100] and [01 0] dendritic columns. Based on the preferential growth criterion of dendrites, a model describing dendritic growth behavior in the laser remelting pool was established, which explains the experimental results well. The results demonstrate that solidification microstructure in the molten pool is influenced by both pool morphology and substrate crystal orientation.

Keywords: laser remelting, single crystal, molten pool, dendrite, transparent model alloy, real-time observation

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Introduction

High-energy beam processing technologies, including laser melting, cladding, welding, and additive manufacturing, all involve the interaction between a high-energy beam and metal to produce a molten pool, followed by rapid cooling and solidification to obtain the final part [1,2]. This means that the solidification microstructure in the molten pool directly determines the final quality of the processed part, making the evolution of microstructure in the molten pool a key research focus in the field of high-energy beam processing [3–7].

Previous studies have shown that molten pool microstructure is affected by both pool morphology and substrate grain orientation. Rappaz et al. [8,9] studied the microstructure of electron beam welded single-crystal Fe-Cr-Ni alloy and established a dendrite growth orientation model in the molten pool using the minimum undercooling principle, deriving the relationship between dendrite growth velocity along the preferred direction and both interface solidification velocity and scanning speed. Yang et al. [10,11] investigated the effect of substrate crystal orientation on the growth direction of laser melting microstructure using DD2 single-crystal alloy, demonstrating that microstructure growth direction in the molten pool is strongly influenced by both substrate grain orientation and laser beam scanning direction. Feng et al. [12,13] performed laser multilayer cladding experiments of Rene95 alloy on the preferred crystal plane of directionally solidified nickel-based superalloy (DD3) and studied the effect of substrate orientation on laser directional solidification microstructure. Liu and Dupont [14,15] established an analytical model and combined it with experiments to investigate the effects of molten pool morphology and substrate grain orientation on dendrite growth in the molten pool. These studies all employed metallographic observation to examine molten pool microstructure. While metallography is a common method for observing microstructure and provides rich information about microstructural features and composition, it can only reveal the final solidified structure, not the dynamic evolution process during solidification. In actual molten pool solidification, local temperature gradient and cooling rate change continuously as the solidification interface advances, meaning the dynamic evolution process of solidification microstructure must significantly influence the final microstructural characteristics. Therefore, researchers have long sought to achieve real-time observation of molten pool solidification processes. However, due to the opacity of metals and the challenging conditions of high temperature, rapid change, and microscopic scale, real-time observation of metal molten pool solidification microstructure has remained a significant challenge.

In recent years, microstructure simulation methods such as phase field (PF), cellular automaton (CA), and Monte Carlo (MC) have been used to simulate solidification microstructure evolution in molten pools under high-energy beam processing. Fallah et al. [16] simulated the laser solid forming process of Ti-Nb alloy, using finite element calculations to obtain the macroscopic temperature distribution in the molten pool and then PF method to analyze the solidification microstructure. Farzadi et al. [17] used PF method to simulate solidification microstructure in Al-Cu alloy TIG weld seams and studied the effect of welding speed on solidification microstructure. Yin et al. [18] combined finite element and CA methods to simulate dendrite growth during laser solid forming. Mishra and Debroy [19] used MC method for three-dimensional microstructure simulation of pure Ti welded joints, quantitatively calculating grain size and distribution morphology. In China, Zhan et al. [20] and Ma [21] have focused on using CA method to simulate solidification microstructure in welding molten pools. Huang et al. [22] and Li et al. [23] also used CA method to simulate weld metal solidification microstructure, reproducing the grain preferred orientation and competitive growth mechanism. Although these simulation efforts have enhanced understanding of solidification microstructure formation in laser molten pools to some extent, the models have undergone significant simplification, and the computational results still require further experimental validation.

Notably, as organic materials with solidification behavior similar to metals, transparent model alloys have been widely used in solidification research due to their transparency and low melting point, which facilitate real-time observation. As early as 1972, Savage and Hrubec [24] pioneered the use of transparent organic camphene to study welding molten pool shape and microstructure evolution. In 2003, Trivedi et al. [25] used a 5 W CO_2 laser to scan a quasi-two-dimensional succinonitrile-1.2% (mass fraction) acetone model alloy sample, real-time observing the morphology and solidification microstructure evolution of the laser molten pool and observing defect formation during solidification. This demonstrates that transparent model alloys can serve as an effective means for real-time evolution studies of laser molten pool solidification. However, their research focused on macro-micro evolution of molten pools on polycrystalline substrates, making it difficult to accurately clarify the effects of substrate orientation and scanning method on molten pool solidification microstructure. To address these issues, this work employs succinonitrile-2.0% (mass fraction) ethanol transparent model alloy to conduct real-time observation of non-steady solidification evolution in laser molten pools, examining the effect of laser scanning direction on the non-steady dendrite growth process in molten pools on single-crystal substrates, with the aim of clarifying the influence 规律 of substrate crystal orientation on molten pool solidification microstructure.

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1 Experimental Methods

Succinonitrile-2.0% (mass fraction) ethanol (SCN-2.0%Eth) transparent model alloy was used as the experimental material. The SCN purity was greater than

99.99%, and the ethanol was analytically pure.

The real-time observation platform for laser remelting molten pools consisted of a laser system, motion system, microscopic observation system, and sample cell, as shown in [Figure 1: see original paper]. The laser system comprised an optical platform, CO_2 laser, mirrors, focusing lens, and related control components, allowing adjustment of laser power and spot size. The motion system consisted of a horizontal translation stage and corresponding control software to move the sample cell relative to the laser head, enabling adjustment of scanning speed and path. The microscopic observation system included an Olympus SZX16 microscope, charge-coupled device (CCD), illumination source, and computer, enabling real-time microscopic observation and image acquisition of the laser remelting process at various magnifications. The sample cell was a quasi-two-dimensional glass box filled with transparent model alloy.

During the experiments, CO_2 laser remelting was performed on SCN-2.0%Eth alloy samples with a thickness of approximately 100 mm in the sample cell. Before the experiment, a SCN-2.0%Eth alloy single crystal was epitaxially grown in the sample cell using a seeding method, with the (001) crystal plane parallel to the sample cell plane and the $\langle 100 \rangle$ crystal direction aligned with the long edge of the sample cell. The seeded sample cell was then held at 40 °C for 12 h to homogenize the alloy.

The experimental parameters were laser power P = 2.0 W and scanning speed v = 0.8 mm/s. By horizontally rotating the sample cell, the angle between the laser scanning direction and the [100] crystal direction of the single-crystal sample was varied. Three scanning directions deviating from the [100] crystal direction by 0°, 20°, and 45° were selected for laser remelting to investigate the effect of single-crystal substrate orientation on dendrite growth evolution behavior in the molten pool.

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2 Results

2.1 Unsteady Evolution Process of Molten Pool

[Figure 2: see original paper] shows the macroscopic morphology evolution of the molten pool during laser remelting along the [100] crystal direction of the SCN-2.0%Eth single crystal at P = 2.0 W and v = 0.8 mm/s, where time t is measured from the start of laser scanning. It can be seen that as laser scanning proceeds, the molten pool morphology transforms from circular to elliptical and finally evolves into a steady teardrop shape. Using the widest point at the side of the molten pool as the boundary, the front interface facing the scanning direction is the melting interface, while the trailing interface is the solidification interface. From the widest point to the tail of the molten pool, the temperature gradient at the solidification interface gradually decreases, and the normal moving velocity gradually increases. The interface morphology evolves through a sequence from planar to cellular to dendritic. The evolution of the solidification interface represents the formation history of molten pool microstructure; therefore, the following sections will focus on examining the evolution behavior of the trailing solidification interface.

2.2 Scanning Direction Parallel to [100] Crystal Orientation

[Figure 3: see original paper] shows the dendrite evolution process in the SCN-2.0%Eth alloy molten pool during laser remelting along the [100] crystal direction of the single-crystal substrate at P = 2.0 W and v = 0.8 mm/s. Initially, upon irradiation, dendritic columns growing along the [100], [010], and [01 0] crystallographic orientations exist at the tail of the elliptical molten pool (Fig. 3a). For simplicity, these three dendritic columns growing along different crystallographic orientations are designated as [100], [010], and [01 0] dendritic columns, respectively. Subsequently, as the molten pool shape evolves toward a steady teardrop shape, continuous competitive elimination occurs among dendritic columns with different primary arm growth directions. Eventually, the [100] dendritic column is completely eliminated, leaving only the [010] and [01 0] dendritic columns growing parallel and symmetrically at both sides of the molten pool (Fig. 3b). The entire non-steady competitive elimination process of molten pool microstructure lasts approximately 10 s. The final steady teardrop-shaped molten pool has a trailing angle of approximately 71°.

2.3 Scanning Direction Deviated by 20° from [100] Crystal Orientation

[Figure 4: see original paper] shows the dendrite evolution process in the SCN-2.0%Eth alloy molten pool during laser remelting at P = 2.0 W and v = 0.8 mm/s along a direction deviated by approximately 20° from the [100] crystal direction of the single-crystal substrate. Initially, at the tail of the elliptical molten pool, [100], [010], and [01 0] dendritic columns also exist (Fig. 4a), but with an asymmetric distribution. During subsequent evolution, the dendrites at the lower side of the molten pool consistently exhibit [01 0] dendritic column growth, while the [100] and [010] dendritic columns at the upper side show competitive growth with alternating appearance. Specifically, when the [100] dendritic column at the upper side grows, its growth direction tilts toward the lower left (Fig. 4b), providing sufficient space for the [010] dendritic column on its upper left to grow (Fig. 4c). When the [010] dendritic column grows, its growth direction tilts toward the lower right (Fig. 4d), allowing the [100] dendritic column to grow again with adequate conditions and space (Figs. 4e and f). Consequently, throughout the scanning process, the [100] and [010] dendritic columns at the upper side of the molten pool maintain alternating growth with a period of approximately 18 s. Simultaneously, the trailing angle of the molten pool changes periodically with this alternating growth: when the [100] dendritic column dominates at the upper side and grows perpendicular to the [01 0] dendritic column at the lower side, the trailing angle increases; when the [010] dendritic column dominates at the upper side and grows relatively parallel to the [01 0] dendritic column at the lower side, the trailing angle decreases. The dendritic morphology after remelting completion (Fig. 4g) shows traces of the competitive alternating growth between the [100] and [010] dendritic columns at the upper side of the molten pool. Additionally, the fusion line between dendrites on both sides shows a wavy pattern with a wavelength of approximately 3 mm. It should be noted that for descriptive convenience, the positional terms (upper/lower) for the molten pool refer to the image orientation; in reality, the sample was placed horizontally, and the molten pool was horizontal, so the upper and lower sides described correspond to the horizontal sides perpendicular to the scanning direction in the actual molten pool.

2.4 Scanning Direction Deviated by 45° from [100] Crystal Orientation

[Figure 5: see original paper] shows the dendrite evolution process in the SCN-2.0%Eth molten pool during laser remelting at P = 2.0 W and v = 0.8 mm/s along a direction deviated by approximately 45° from the [100] crystal direction of the single-crystal substrate. From the initial solidification of the elliptical molten pool, the [100] and [01 0] dendritic columns at both sides of the pool tail grow perpendicular to each other, continuing through to the later steady teardrop-shaped molten pool stage. The two dendritic regions are symmetrically distributed, with a steady trailing angle of 78°.

3 Analysis and Discussion

To describe the microstructure evolution in the molten pool, we first analyze dendrite growth behavior with reference to the pool shape. [Figure 6: see original paper] shows a schematic diagram of a two-dimensional teardrop-shaped molten pool, where the trailing angle is 2 ; the laser scans horizontally from right to left at scanning speed v; the angle between the solidification interface normal and the laser scanning direction is ω; the normal moving velocity of the solidification interface is v ; the (001) crystal plane of the single-crystal substrate is parallel to the quasi-two-dimensional molten pool plane; the scanning direction is deviated by angle from the [100] crystal direction, with counterclockwise deviation defined as positive and clockwise as negative. For the SCN-2.0%Eth alloy with a bcc crystal structure, the $\langle 100 \rangle$ crystal direction is the preferred dendrite growth direction. Therefore, in the quasi-two-dimensional molten pool parallel to the (001) crystal plane, there are four possible dendrite growth directions, designated as U[100], U[010], D[100], and D[01 0].

From geometric relationships, the dendrite growth velocity v[hkl] for [hkl] orientation can be obtained as: v[hkl] = v /cos(ϕ), where ϕ is the angle between the [hkl] orientation and the solidification interface normal. It should be noted that during the non-steady transition from near-circular to teardrop-shaped molten pool morphology, ω continuously changes to at the tail endpoint. When the steady teardrop shape shown in [Figure 6: see original paper] is reached, ω is approximately $\pi/2$ - . Therefore, in a steady molten pool, the dendrite growth velocity is constant, with $\omega = \pi/2$ - .

The molten pool trailing angle   always lies within the range $(0, \pi)$, meaning $(0, \pi)$. Meanwhile, crystal orientation symmetry determines that $(-\pi/4, \pi/4)$. To clearly analyze the effects of   and   on dendrite growth in the molten pool, we establish a rectangular coordinate system as shown in [Figure 7: see original paper], dividing the rectangular region into four zones: a, b, c, and d.

Zone a: When $\cos(\pi/2 -\ \ +\ \ ) > \cos(\pi/2 -\ \ -\ \ )$, we have U[100] > D[100] and U[010] > D[01 0]. The dendrite with the smallest growth velocity has the smallest tip undercooling, positioning its tip ahead of other oriented dendrites and thus placing it in the most favorable position during growth competition—this is the so-called preferential growth criterion. According to this criterion, when the molten pool reaches steady state, both sides will show parallel relative growth of [010] and [01 0] dendritic columns.

Zone b: When $\cos(\pi/2 -\ \ +\ \ ) < \cos(\pi/2 -\ \ -\ \ )$, we have U[100] < D[100] and U[010] > D[01 0]. According to the preferential growth criterion, at steady state the upper side will show [100] dendrites and the lower side [01 0] dendrites, growing perpendicular to each other.

Zone c: When $\cos(\pi/2 -\ \ +\ \ ) > \cos(\pi/2 -\ \ -\ \ )$, we have U[100] > D[100] and U[010] < D[01 0]. In this zone, the preferred growth direction on both sides of the molten pool is [100].

Zone d: When $\cos(\pi/2 -\ \ +\ \ ) < \cos(\pi/2 -\ \ -\ \ )$, we have U[100] < D[100] and U[010] < D[01 0]. According to the preferential growth criterion, at steady state the upper side will show [010] dendrites and the lower side [100] dendrites, growing perpendicular to each other.

Thus, when the molten pool reaches steady state, dendrites on both sides grow parallel and opposite to each other in zones a and c, while they grow perpendicular to each other in zones b and d.

We now apply this dendrite growth model to discuss the three experimental results:

1. **When scanning direction is parallel to [100] crystal direction:** From [Figure 3: see original paper], during non-steady evolution of the molten pool, the trailing angle   changes from 180° to 71°, meaning   decreases from 90° to 35.5°. According to [Figure 7: see original paper], this corresponds to the molten pool evolution passing through zone c and finally stabilizing in zone a. Zone c features [100] dendritic column growth, while zone a features parallel relative growth of [010] and [01 0] dendritic columns. This is consistent with the experimental observation that [100] dendritic columns are gradually eliminated, leaving [010] and [01 0] dendritic columns growing symmetrically.
2. **When scanning direction deviates by ~20° from [100] crystal di-**

rection: As shown in [Figure 4: see original paper], throughout the evolution process, the molten pool exhibits asymmetric dendrite growth, with competitive alternating growth between [100] and [010] dendritic columns on one side, accompanied by periodic changes in the molten pool trailing angle and fluctuations in the fusion line. As illustrated in [Figure 7: see original paper], this corresponds to the dendrite evolution at the molten pool tail passing through zone c and finally oscillating between zones a and b. This occurs because the steady-state trailing angle depends on process parameters such as laser power and scanning speed, and under the present experimental conditions, the trailing angle happens to be located in the critical region between zones a and b, creating an unstable state that causes the dendritic column growth direction in the molten pool to oscillate between these two zones.

3. **When scanning direction deviates by ~45° from [100] crystal direction:** From [Figure 5: see original paper], during non-steady evolution of the molten pool, the trailing angle changes from 180° to 78°, meaning decreases from 90° to 39°. According to [Figure 7: see original paper], the molten pool remains in zone b throughout the process, where [100] and [01 0] dendritic columns grow perpendicular to each other. Experimentally, it can be observed that the molten pool grows with perpendicular dendritic columns on both sides from the beginning until the teardrop shape is reached, maintaining this perpendicular growth pattern throughout the entire evolution process.

4 Conclusions

1. Throughout the laser remelting process, the macroscopic morphology of the molten pool transforms from circular to elliptical and finally evolves into a steady teardrop shape. Using the widest point at the side of the molten pool as the boundary, the front interface facing the scanning direction is the melting interface, while the trailing interface is the solidification interface. From the widest point to the tail of the molten pool, the solidification interface morphology evolves through a sequence from planar to cellular to dendritic.

2. When laser scanning is performed along the [100] crystal direction of the single-crystal substrate, [100] dendritic columns at the molten pool tail compete with [010] and [01 0] dendritic columns. The [100] dendritic columns are gradually eliminated, finally evolving into parallel symmetric growth of [010] and [01 0] dendritic columns. When the scanning direction deviates by 20° from the [100] crystal direction, the entire molten pool exhibits asymmetric growth: one side consistently shows [01 0] dendritic column growth, while the other side shows competitive alternating growth between [100] and [010] dendritic columns, accompanied by peri-

odic changes in the molten pool trailing angle and periodic fluctuations in the fusion line between dendrites on both sides. When the scanning direction deviates by 45° from the [100] crystal direction, the molten pool consistently exhibits perpendicular growth between [100] and [01 0] dendritic columns.

3. Based on the preferential growth criterion, a model describing dendrite growth behavior in the molten pool was established, which explains the experimental results well. The results demonstrate that solidification microstructure in the molten pool is jointly influenced by molten pool morphology and substrate crystal orientation.

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