Real-time Observation of Microstructure in Laser Remelting Molten Pool (Postprint)

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Abstract

Using the transparent model alloy succinonitrile-2.0% (mass fraction) ethanol (SCN-2.0%Eth), the evolution of dendrite growth in the melt pool was observed in real time during laser remelting on the (001) crystal plane of a single-crystal substrate, with the scanning direction deviating from the substrate [100] crystal orientation at different angles. It was found that when scanning along a direction parallel to the [100] crystal orientation, the melt pool eventually evolved into parallel symmetric growth of [010] dendrite arrays and dendrite arrays; when the angle between the scanning direction and the [100] crystal orientation was 20°, the entire melt pool exhibited an asymmetric oscillatory growth mode, i.e., one side consistently showed dendrite array growth, while the other side showed alternating competitive growth between [100] and [010] dendrite arrays; when the angle between the scanning direction and the [100] crystal orientation reached 45°, the melt pool consistently exhibited perpendicular growth of [100] dendrite arrays and dendrite arrays. Based on the dendrite preferred growth criterion, a model describing dendrite growth behavior in the melt pool was established, which well explained the experimental results. The results indicate that the solidification microstructure in the melt pool is jointly influenced by the melt pool morphology and the substrate crystal orientation.

Full Text

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 processing technologies (including laser, electron beam, and arc) 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 temperatures, rapid solidification, and material opacity. In this study, using a transparent model alloy of succinonitrile-2.0% ethanol (SCN-2.0%Eth) (mass fraction), we conducted real-time observations of solidification microstructure evolution in the molten pool during laser surface remelting of a single-crystal substrate (001) crystal plane, with the laser scanning direction deviating at various angles from the substrate [100] crystal orientation.

The experimental results revealed three distinct growth modes: When scanning parallel to the [100] crystal direction, [010] dendritic columns grew symmetrically in the molten pool. When the scanning direction deviated by 20° from the [100] crystal orientation, asymmetric oscillatory growth occurred, with one side consistently exhibiting dendritic column growth while [100] and [010] dendritic columns competed and alternated on the other side. When the deviation angle reached 45°, the molten pool consistently showed perpendicular growth of [100] and [010] dendritic columns.

Based on the preferential growth criterion for dendrites, we established a model describing dendritic growth behavior in laser remelting pools that successfully explains the experimental observations. The results demonstrate that solidification microstructure formation in laser remelting pools is jointly influenced by pool morphology and substrate crystal orientation.

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

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

Previous studies have shown that molten pool microstructure is influenced by both pool morphology and substrate grain orientation. Rappaz et al. [8,9] investigated the microstructure of electron beam welded single-crystal Fe-Cr-Ni alloy and established a dendrite growth orientation model based on the minimum undercooling principle, relating dendrite growth velocity along preferred directions to interface solidification velocity and scanning speed. Yang et al. [10,11] studied the effect of substrate crystal orientation on laser melting microstructure growth directions using DD2 single-crystal alloy, demonstrating that microstructure growth directions are strongly affected by both substrate grain orientation and laser beam scanning direction. Feng et al. [12,13] performed laser multi-layer cladding of Rene95 alloy on the preferred crystal plane of directionally solidified nickel-based superalloy DD3 and investigated substrate orientation effects on laser-directed solidification microstructure. Liu and Dupont [14,15] developed an analytical model combined with experiments to study the influence of pool morphology and substrate grain orientation on dendritic growth within molten pools. These studies all employed metallographic observation methods, which are widely used for microstructure analysis and provide rich information about microstructural features and composition. However, metallography can only reveal the final solidified structure, not the dynamic evolution during solidification. During actual molten pool solidification, local temperature gradients and cooling rates change continuously as the solidification front advances, making the dynamic evolution process crucial to the final microstructural characteristics. Therefore, researchers have long sought to achieve real-time observation of molten pool solidification. Nevertheless, due to metal opacity and the challenging conditions of high temperature, rapid change, and microscopic scales, real-time observation of metallic molten pool solidification remains difficult.

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

Notably, transparent model alloys, as organic analogs of metals with similar solidification behavior, have been widely used in solidification research due to their transparency and low melting points, which facilitate real-time observation. As early as 1972, Savage and Hrubec [24] pioneered the use of transparent organic camphene to study welding pool shape and microstructure evolution. In 2003, Trivedi et al. [25] used a 5 W CO laser to scan quasi-two-dimensional succinonitrile-1.2% acetone model alloy specimens, real-time observing molten pool morphology and solidification microstructure evolution, including defect formation during solidification. These studies demonstrate that transparent model alloys are valuable tools for investigating real-time evolution of laser molten pool solidification. However, their research focused on macro- and micro-evolution of pools on polycrystalline substrates, making it difficult to clearly elucidate substrate orientation and scanning mode effects on solidification microstructure. To address these issues, this work employs succinonitrile-2.0% ethanol transparent model alloy to conduct real-time observations of non-steady solidification evolution in laser molten pools, investigating the influence of laser scanning direction on non-steady dendritic growth processes in single-crystal substrates to clarify the role of substrate crystal orientation on solidification microstructure formation.

Experimental Methods

The experimental material was succinonitrile-2.0% ethanol (SCN-2.0%Eth) transparent model alloy. The SCN purity was greater than 99.99%, and the ethanol was analytical grade.

The real-time observation platform for laser remelting molten pools consisted of a laser system, motion system, microscopic observation system, and specimen cell, as shown in [Figure 1: see original paper]. The laser system comprised an optical platform, CO laser, mirrors, focusing lens, and control components, enabling adjustment of laser power and spot size. The motion system consisted of a horizontal translation stage and control software to move the specimen cell relative to the laser head, allowing control 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 at various magnifications. The specimen cell was a quasi-two-dimensional glass container filled with transparent model alloy.

During experiments, CO laser remelting was performed on SCN-2.0%Eth alloy specimens approximately 100 m thick in the specimen cell. Prior to experiments, a SCN-2.0%Eth single crystal was epitaxially grown in the specimen cell using a seeding method, with the (001) crystal plane parallel to the cell plane and the <100> crystal direction aligned with the cell' s long axis. The seeded specimen cell was then held at 40 °C for 12 hours 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 specimen cell, the angle between the laser scanning direction and the [100] crystal direction of the single-crystal specimen was varied. Three scanning directions deviating 0°, 20°, and 45° from the [100] crystal direction were selected for laser remelting to investigate the influence of single-crystal substrate orientation on dendritic growth evolution behavior in the molten pool.

Results and Discussion

2.1 Non-steady Evolution of Molten Pool [Figure 2: see original paper] shows the non-steady macroscopic morphology evolution of the molten pool during laser remelting along the [100] crystal direction of SCN-2.0%Eth single-crystal alloy at P = 2.0 W and v = 0.8 mm/s, where time t is measured from the start of laser scanning. As laser scanning progresses, the pool morphology transforms from circular to elliptical, and finally evolves into a steady teardrop shape. Using the widest point at the pool side as a 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 pool tail, the temperature gradient at the solidification interface gradually decreases while the normal interface velocity increases from zero, causing the interface morphology to evolve from planar to cellular and then to dendritic. The evolution of the solidification interface represents the formation history of the pool microstructure; therefore, subsequent analysis focuses on the evolution behavior of the trailing solidification interface.

2.2 Scanning Direction Parallel to [100] Crystal Orientation [Figure 3: see original paper] illustrates the dendritic 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, during irradiation, dendritic columns growing along three crystallographic orientations ([100], [010], and [010]) exist at the tail of the elliptical molten pool ([Figure 3: see original paper]a). For simplicity, these three dendritic columns growing along different crystallographic orientations are designated as [100], [010], and [010] dendritic columns. Subsequently, as the pool shape evolves toward a steady teardrop morphology, continuous competitive elimination occurs among dendritic columns with different primary arm growth directions. Eventually, the [100] dendritic columns are completely eliminated, leaving only parallel symmetric growth of [010] and [010] dendritic columns on both sides of the pool ([Figure 3: see original paper]b). The entire non-steady competitive elimination process lasts approximately 10 seconds. The final steady teardrop-shaped pool has a trailing angle of about 71°.

2.3 Scanning Direction Deviating 20° from [100] Crystal Orientation [Figure 4: see original paper] presents the dendritic evolution process in the SCN-2.0%Eth alloy molten pool during laser remelting along a direction deviating approximately 20° from the [100] crystal direction at P = 2.0 W and v = 0.8 mm/s. Initially, [100], [010], and [010] dendritic columns also exist at the tail of the elliptical pool ([Figure 4: see original paper]a), but with an asymmetric distribution. During subsequent evolution, the lower side of the pool consistently exhibits [010] dendritic columns, while competitive growth occurs between [100] and [010] dendritic columns on the upper side, with alternating dominance. Specifically, when [100] dendritic columns grow on the upper side, their growth direction tilts left-downward ([Figure 4: see original paper]b), providing adequate space for [010] dendritic columns to grow on the upper-left side ([Figure 4: see original paper]c). When [010] dendritic columns grow, their right-downward tilt ([Figure 4: see original paper]d) creates favorable conditions and space for [100] dendritic column growth ([Figure 4: see original paper]e and f). Consequently, throughout the scanning process, [100] and [010] dendritic columns on the upper side continue alternating with a period of approximately 18 seconds. This alternating growth is accompanied by periodic variation of the pool trailing angle: when [100] dendritic columns dominate on the upper side and grow perpendicular to the lower [010] dendritic columns, the trailing angle increases; when [010] dendritic columns dominate and grow parallel to the lower [010] dendrites, the trailing angle decreases. The final solidified dendritic morphology ([Figure 4: see original paper]g) reveals traces of the competitive alternating growth between [100] and [010] dendritic columns on the upper side, with the fusion line between both sides showing wavy patterns with a wavelength of about 3 mm. For descriptive convenience, the pool orientation references the image position; in reality, the specimen and pool are horizontally positioned, so the described upper and lower sides correspond to the horizontal sides perpendicular to the scanning direction.

2.4 Scanning Direction Deviating 45° from [100] Crystal Orientation

[Figure 5: see original paper] shows the dendritic evolution process in the SCN-2.0%Eth molten pool during laser remelting along a direction deviating approximately 45° from the [100] crystal direction at P = 2.0 W and v = 0.8 mm/s. From the initial elliptical pool stage, [100] and [010] dendritic columns grow perpendicular to each other on both sides of the pool tail, continuing through to the later steady teardrop stage. The two dendritic zones are symmetrically distributed, with a steady-state trailing angle of [010].

Theoretical Analysis

To describe microstructure evolution in the molten pool, we first analyze dendritic growth behavior with reference to pool shape. [Figure 6: see original paper] shows a schematic of a two-dimensional teardrop-shaped molten pool, where is the pool trailing angle, v is the horizontal laser scanning speed (right to left), w is the angle between the scanning direction and solidification interface normal, v_i is the normal velocity of the solidification interface, is the angle between the scanning direction and the [100] crystal direction (positive for counterclockwise deviation, negative for clockwise), and the (001) crystal plane of the single-crystal substrate is parallel to the quasi-two-dimensional pool plane. For SCN-2.0%Eth alloy with a bcc crystal structure, the <100> directions are the preferred dendritic growth directions, resulting in four possible dendritic growth directions within the quasi-two-dimensional pool parallel to the (001) plane.

The geometric relationships yield the following velocity components for dendrites with [hkl] orientation:

$$v_{[hkl]} = v_i \cos(\varphi_{[hkl]})$$

where $v_{[hkl]}$ is the growth velocity of dendrites with [hkl] orientation and $\varphi_{[hkl]}$ is the angle between the [hkl] orientation and the solidification interface normal. During the non-steady transition from near-circular to teardrop shape, w varies continuously from $\pi/2$ at the widest point to $\pi/2 - \theta$ at the tail endpoint when reaching the steady teardrop shape shown in [Figure 6: see original paper]. Therefore, the dendritic growth velocities in the steady-state pool are:

$$v_U^{[100]} = v \cos(\theta + \phi)$$
$$v_U^{[010]} = v \cos(\theta - \phi)$$
$$v_D^{[100]} = v \cos(\theta - \phi)$$
$$v_D^{[010]} = v \cos(\theta + \phi)$$

The pool trailing angle always falls within the range $0 \leq \theta \leq \pi$, meaning $\theta \in [0, \pi]$. The symmetry of crystal orientation determines that $\phi \in [-\pi/4, \pi/4]$. To clearly analyze the influence of θ and ϕ on dendritic growth in the pool, a rectangular coordinate system was established as shown in [Figure 7: see original paper].

Two lines divide the rectangular region into four zones (a, b, c, and d) for analysis:

Zone a: $\sin(\theta + \phi) > \cos(\theta + \phi)$ and $\sin(\theta - \phi) > \cos(\theta - \phi)$. According to the preferential growth criterion, dendrites with minimum growth velocity have the smallest tip undercooling and advance ahead of others, placing them in the most favorable competitive position. In this zone, the steady-state pool exhibits parallel opposite growth of [010] and [010] dendrites on both sides.

Zone b: $\sin(\theta + \phi) < \cos(\theta + \phi)$ and $\sin(\theta - \phi) > \cos(\theta - \phi)$. Based on the preferential growth criterion, the steady-state pool shows [100] dendrites on the upper side and [010] dendrites on the lower side, growing perpendicular to each other.

Zone c: $\sin(\theta + \phi) < \cos(\theta - \phi)$ and $\sin(\theta - \phi) < \cos(\theta - \phi)$. In this zone, both sides of the pool have [100] as the preferential growth direction.

Zone d: $\sin(\theta + \phi) > \cos(\theta + \phi)$ and $\sin(\theta - \phi) < \cos(\theta - \phi)$. According to the preferential growth criterion, the steady-state pool exhibits [010] dendrites on the upper side and [100] dendrites on the lower side, growing perpendicular to each other.

Thus, in steady-state pools, zones a and c show parallel opposite dendritic growth on both sides, while zones b and d show perpendicular dendritic growth.

The experimental results can now be discussed using this dendritic growth model:

1. **Scanning direction parallel to [100] crystal direction:** [Figure 3: see original paper] shows that during non-steady evolution, the trailing angle θ decreases from 180° to 71°, meaning θ changes from 90° to 36°. According to [Figure 7: see original paper], this corresponds to the pool evolution passing through zone c and finally stabilizing in zone a. Zone c features [100] dendritic column growth, while zone a exhibits parallel opposite growth of [010] and [010] dendritic columns. This matches the experimental observation of [100] dendritic columns being gradually eliminated, leaving symmetric [010] and [010] dendritic columns.
2. **Scanning direction deviating ~20° from [100] crystal direction:** [Figure 4: see original paper] shows asymmetric dendritic growth throughout evolution, with competitive alternating growth between [100] and [010] dendritic columns on one side, accompanied by periodic variation of the trailing angle and undulation of the fusion line. As shown in [Figure 7: see original paper], this corresponds to the dendritic evolution at the pool tail passing through zone c and oscillating between zones a and b. Since the steady-state trailing angle depends on process parameters such as laser power and scanning speed, the trailing angle under these experimental conditions lies precisely at the critical region between zones a and b, creating an unstable state that causes oscillation of dendritic growth directions between the two zones.
3. **Scanning direction deviating ~45° from [100] crystal direction:** [Figure 5: see original paper] shows that during non-steady evolution, the trailing angle θ decreases from 180° to 78°, meaning θ changes from 90° to 39°. According to [Figure 7: see original paper], the pool remains in zone b throughout, where [100] and [010] dendritic columns grow perpendicular to each other. Experimentally, the pool exhibited perpendicular dendritic columns on both sides from the beginning through to the teardrop shape formation.

Conclusions

This study investigated non-steady solidification evolution in laser remelting pools using succinonitrile-2.0% ethanol transparent model alloy, focusing on the effects of laser scanning direction on non-steady dendritic growth in single-crystal substrates. The main findings are:

1. Throughout laser remelting, the macroscopic pool morphology evolves from circular to elliptical and finally to a steady teardrop shape. Using the widest point as a 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 pool tail, the solidification interface morphology evolves from planar to cellular to dendritic.

2. When laser scanning is performed along the single-crystal substrate [100] direction, competitive growth occurs among [100], [010], and [010] dendritic columns at the pool tail, with [100] dendritic columns being gradually eliminated, resulting in final parallel symmetric growth of [010] and [010] dendritic columns. When the scanning direction deviates 20° from the [100] direction, the pool exhibits asymmetric growth: one side consistently shows [010] dendritic columns while the other side displays alternating competitive growth between [100] and [010] dendritic columns, accompanied by periodic variation of the pool trailing angle and undulation of the fusion line. At a deviation angle of 45°, the pool consistently shows perpendicular growth of [100] and [010] dendritic columns.
3. Based on the preferential growth criterion, a model describing dendritic growth behavior in molten pools was established that successfully explains the experimental results, demonstrating that solidification microstructure formation is jointly influenced by pool morphology and substrate crystal orientation.

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