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Microstructure evolution and tensile property of a first-generation single crystal superalloy fabricated by laser melting deposition

Guowei Wang\textsuperscript{1,2} \textcopyright, Xianfeng Shen\textsuperscript{1}, Jialin Yang\textsuperscript{1}, Jingjing Liang\textsuperscript{2,3}, Yizhou Zhou\textsuperscript{2,3} and Xiaofeng Sun\textsuperscript{2}

\textsuperscript{1} Institute of Machinery Manufacturing Technology, China Academy of Engineering Physics, Mianyang 621900, People’s Republic of China
\textsuperscript{2} Institute of Metal Research, Chinese Academy of Sciences, Shenyang 110016, People’s Republic of China
\textsuperscript{3} Authors to whom any correspondence should be addressed.

E-mail: jjliang@imr.ac.cn and yzzhou@imr.ac.cn

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Abstract

Additive manufacturing (AM) of single crystal superalloys has got some progress in recent researches, but there are few reports on eliminating recrystallization or mechanical properties of single crystal superalloys by AM. In this work, single-crystal samples of SRR99 were fabricated at high temperature by laser melting deposition (LMD). Owing to the high temperature of substrate in the deposition process, recrystallization in the following heat treatment process was eliminated. The contrast sample of SRR99 was prepared by directional solidification, and the microstructure evolution during heat treatment and tensile property of deposited samples were analyzed. The results showed that the shapes of the $\gamma'$ phase became irregular after solution treatment in deposited samples. After a solid solution and aging treatment, the $\gamma'$ phase size is larger and the $\gamma'$ volume fraction is slightly lower in deposited samples than in the contrast sample. As a result, the yield and tensile strength of deposited samples are slightly lower than that of contrast samples, but the plasticity of deposited samples is better.

1. Introduction

Additive manufacturing has the potential to directly produce single-crystal blade with complex structure [1–3], instead of preparing wax pattern or ceramic shell [4, 5]. But the research on AM of single crystal superalloys is still in a primary stage, much experimental and theoretical research has to be done [6, 7].

To understand the dendritic growth law in additive manufacturing, laser remelting experiments of single crystal superalloys were usually used [8–10]. In the remelted microstructures, most of the dendrites followed the law of epitaxial growth except for some stray grains [11, 12]. However, the dendrites epitaxial growth directions from different temperature gradient directions are different [13, 14], such as the researches of laser scanning along [100] orientation on the (001) surface [15, 16]: the dendrites grow along [001] at the bottom of molten pool, the dendrites grow along [100] at the middle-upper part, and the dendrites grow along [010] or [0\overline{1}0] at the left side and right side. It can be concluded that the growth law of dendrites can be traced in the remelting microstructure of single crystal superalloys.

The microstructure of laser melting deposition on a single crystal substrate is similar to that of the laser remelting sample. In the single-pass deposition experiment of laser scanning along [100] orientation on the (001) surface [17–19], the most of dendrites grow along [001] or [100], and only a small amount of dendrites grow along other orientations. Based on the results of single-pass deposition, multi-layer deposition of single-crystal structure can be obtained through appropriate process parameters [20, 21]. In the past works [22, 23], the cross-section of a single-crystal structure by laser melting deposition is generally less than 10 mm$^2$, and it is not suitable for the tensile experiment. Except for laser melting deposition, Ni-base single crystal superalloy was also produced by selective electron beam melting [24]. The cross-section of the single-crystal structure of more than 10 mm$^2$ was made out, but the cracks are yet to be overcome [24–26]. Furthermore, there are some other difficulties to prepare a large-sized single crystal superalloy [27–30], such as control of epitaxial growth, recrystallization and, etc.
In this paper, control of epitaxial growth in large size was achieved based on the laser epitaxial growth deposition model \[31, 32\]. In-situ preheating technology was used to keep the high temperature of substrate, thus, the crack in deposition and recrystallization in heat treatment were eliminated. Additionally, the tensile property of deposited single crystal superalloy after heat treatment was measured.

2. Experiments

2.1. Laser melting deposition of single crystal superalloy SRR99

The alloy SRR99 used in this study is a first-generation single crystal superalloy. The nominal chemical compositions of the alloy in weight percent are given in table 1. A single crystal superalloy plate with a dimension of \(21 \times 11 \times 10\) mm was used as the substrate, and the substrate was solution heat-treated. The argon atomized powders (75–150 \(\mu\)m) were used as the deposited material.

Experiments of laser melting deposition were conducted using a CO\(_2\) laser materials processing system with a computer-numerical-controlled (CNC) work station. Argon gas was used as a powder carrier gas to prevent the melt-pool from oxidization. An in situ warm-up process was carried out to raise the substrate temperature before deposition. The in situ warm-up parameters were as follows: laser power setting at 1600 W, beam diameter about 4 mm, laser scanning speed 1000 mm min\(^{-1}\) and no powder. After the warm-up, the LMD process was conducted as following parameters: laser power setting at 1600 W, beam diameter about 2 mm, laser scanning speed 1200 mm min\(^{-1}\) and powder feeding rate about 7 g min\(^{-1}\).

In the LMD process, the laser scanning paths were shown in figure 1. In the 1st, 5th, 9th and more layers of deposition, the cladding strategy worked as Track 1. Similarly, Track 2, 3, 4 corresponded to the 2nd, 3rd, 4th and more cladding layers, respectively. Based on the deposition methods as above, a single crystal superalloy plate was deposited as figure 2. Its dimension is about \(18 \times 10 \times 23\) mm. To study the microstructure and tensile property of deposited sample (sample A), a plate (sample B) as comparison was directionally solidified into [001] single crystal by Bridgman withdraw technique. The measured chemical compositions of samples A and B were listed in table 1.

2.2. Heat treatment and characterization

Then, deposition sample A and casting sample B were heat-treated by using common parameters for the alloy. Sample A1 and B1 represented no heat-treat, sample A2 and B2 (solution treatment), sample A3 and B3 (solution and aging treatment). The corresponding heat-treat conditions of these samples were listed in table 2.

![Figure 1. Schematic diagram of laser scanning paths during laser deposition.](image)

| Table 1. The nominal composition and measured composition of experimental alloys. |
|------------------|-----|-----|-----|-----|-----|-----|
|                  | Ni  | Cr  | Co  | W   | Al  | Ti  | Ta  |
| Nominal composition (in wt%) | Bal. | 8.00 | 5.00 | 10.00 | 5.50 | 2.20 | 3.00 |
| Sample A (deposition) (in wt%) | Bal. | 7.99 | 5.07 | 9.62 | 5.16 | 2.10 | 2.84 |
| Sample B (casting) (in wt%) | Bal. | 7.87 | 5.08 | 9.66 | 5.44 | 2.09 | 2.93 |
| Sample A (deposition) (in at%) | Bal. | 9.10 | 5.10 | 3.10 | 11.33 | 2.60 | 0.93 |
| Sample B (casting) (in at%) | Bal. | 8.95 | 5.10 | 3.11 | 11.92 | 2.58 | 0.96 |

In this paper, control of epitaxial growth in large size was achieved based on the laser epitaxial growth deposition model \[31, 32\]. In-situ preheating technology was used to keep the high temperature of substrate, thus, the crack in deposition and recrystallization in heat treatment were eliminated. Additionally, the tensile property of deposited single crystal superalloy after heat treatment was measured.
Metallographic samples were machined by wire electric discharging cutting, followed by standard mechanical grinding and polishing procedures. Before chemically etched, samples A1 and B1 were taken photos to observe microporosity by optical microscopy (OM), and microporosity was measured by image binarization. Then, they were etched in a mixed solution (10 g CuSO₄ + 50 ml HCl + 40 ml H₂O) about 7 s for metallograph. The first dendritic spacing was counted through three metallographic pictures. Furthermore, they were etched in the above solution for about 15 s for scanning electron microscopy (SEM). The crystallographic orientation [100] of the samples was observed. Three SEM pictures were taken into statistics for the size and volume fraction of γ′.

The tensile tests at room temperature were evaluated for samples A3 and B3 on an Instron 5848 tensile testing machine. Figure 3 indicates the size and section shape of the tensile test specimen. Five samples were tested for each case (tensile along [001] direction). The tensile fracture surfaces of A3 and B3 were examined by SEM.

3. Results

3.1. Microporosity and dendritic morphology
OM was employed to reveal the microstructure of deposited sample A1 and the result is shown in figure 4. The metallograph depicts that the dendrites are growing along [001] direction and there is no crack in the deposits. According to the researches [20, 30] about epitaxial growth, the single crystal superalloy plate has been prepared successfully by LMD.

Microporosity in casting single crystal superalloy is a common defect. Therefore, it is necessary to measure the microporosity to appraise the quality of deposition. Figure 5 shows the microporosity in the deposited sample A3 and casting sample B3. The area percentages of microporosity are further measured, their mean values of samples A and B are 0.24% and 0.26%, respectively.

Comparing with the casting sample, the deposition plate has a finer rapidly solidified cellular-dendrite structure which has little features of secondary dendrite arms as figures 6(a) and (b). And the average measured
values of first dendritic spacing of sample A1 and B1 are about 43 and 480 μm, respectively. After solution treatment, the dendritic morphology in sample A2 has faded as figure 6(c), while the dendritic morphology can be seen in sample B2. And there is no recrystallization after solution treatment in sample A2. Moreover, dendritic morphology can’t be seen in sample A3 after solution and aging treatment, and no recrystallization as figure 6(e). In contrast, dendritic morphology can be seen in sample B3.

3.2. Distribution and size of γ′ phase
The high-magnification SEM micrographs shown in figure 7 depicts that the sizes of γ′ are different in different regions or by different preparation technologies. The γ′ sizes of A1 and B1 are listed in table 3. Apparently, in
both deposition and casting samples, the size of $\gamma'$ in the dendritic arm regions was smaller than that in the interdendritic zones. The value of $\gamma'$ size from small to large order is dendritic A1, interdendritic A1, dendritic B1, and interdendritic B1.

After the solution or aging treatment, the difference of $\gamma'$ size in different regions is unremarkable. And the following results will not distinguish dendritic from interdendritic zones. Morphologies and distribution of $\gamma'$ phase in samples A2, B2, A3, and B3 are shown in figure 8. The mean values of $\gamma'$ size in these samples are measured and shown in table 4. Histogram of the measured size of $\gamma'$ precipitates was plotted in figure 9. The measured values of $\gamma'$ volume fraction are 67.6% and 70.4% in sample A3 and B3, respectively.

3.3. Mechanical properties

The tensile properties (yield strength (YS), ultimate tensile strength (UTS), elongation at break) of samples A3 and B3 are given in table 5. Results show that the tensile and yield strength of sample A3 is smaller than that of B3. But the ductility of sample A3 is apparently higher than it of B3. After the tensile test, the results of the SEM fractographic study of samples A3 and B3 are revealed in figure 10. Both deposition and casting samples showed a typical cleavage fracture. The size of the tear ridge in sample A3 is finer and denser than B3. Moreover, the plastic deformation around the micropore of sample A3 is typically larger than B3.

4. Discussion

4.1. Laser deposition principle of single crystal superalloy

In a single crystal weld, geometrical analysis [13] and vectorization method [14] were carried out to predict the dendrites growth direction and relate the dendrite growth velocities in the different (100) directions to the weld velocities. Based on these calculation models, some works [11] have predicted the tendency and distribution of stray grains in the laser weld. In the work of Wang et al [32], section 4.2 has given out the deposition principle which can ensure [001] dendrites epitaxial growth and eliminate stray grains by remelting in the next layer deposition.

Furthermore, the effects of the flowing field cannot be neglected in the deposition of single crystal superalloy. The early studies [33] have shown that the flowing field will lead to the dendrites deflect toward an upstream direction. In the recent work [30], a variation of crystal orientation during epitaxial growth of dendrites by laser deposition owing to flowing field was reported. Instead of stray grains, it is a new phenomenon that will lead to
the failure of single-crystal deposition. And the control sample in the work depicts that variation of crystal orientation can be eliminated by selected suitable parameters.

According to the above analyze, a single crystal deposition can be produced as figure 4, the dendrites are vertical and grow along a straight line in [001] orientation. However, recrystallization is inevitable if the deposition has higher levels of residual stress. And so, the in situ preheating was carried out before deposition which can heat the substrate to red-hot temperature. Besides, the high power of the laser was used to ensure the
Figure 7. SEM micrographs showing difference of $\gamma'$ in the dendritic arm (a), (b) and the interdendritic regions (c), (d) of different samples: (a), (c) of A1; (b), (d) of B1.

Figure 8. SEM micrographs showing difference of $\gamma'$ morphology in different samples: (a) A2; (b) B2; (c) A3; (d) B3.
deposition keep red-hot temperature. Finally, the deposition with lower levels of residual stress has no recrystallization after heat treatment as figure 6.

4.2. The microstructure differences between deposition and casting

It is well known that the primary dendrite arm spacing value \[10\] is the result of the temperature gradient and solidification cooling rate. Because of the high-temperature gradient and rapid solidification cooling rate, the dendrite arm spacing value in the deposition is about ten percent of it in directional solidification casting, and the secondary dendrite arm is greatly suppressed in the deposition. Also, the \(\gamma'\) size in sample A1 is less than half of it in sample B1. The difference of \(\gamma'\) size in sample A1 between dendritic and interdendritic is smaller than in sample B1.

In this work, the same heat treatment condition was used for deposition and casting. The size of \(\gamma'\) in the deposition is bigger after the solution, and the shape of \(\gamma'\) is irregular. It failed to achieve the aim of getting a fine and homogeneous \(\gamma'\) phase by solution treatment. After the solution and aging treatment, the \(\gamma'\) size in the deposition is bigger than it in casting. One possible reason is that the irregular size of \(\gamma'\) leads to bigger driving force of \(\gamma'\) growth. And so, a suitable solution heat-treat parameter is needed which may be higher solution temperature or the solution treatment is not essential.

**Table 4.** Measurements of average \(\gamma'\) size of samples after heat-treatment.

| Sample name | A2  | B2  | A3  | B3  |
|-------------|-----|-----|-----|-----|
| \(\gamma'\) size (nm) | 265 | 263 | 396 | 348 |

**Table 5.** Room temperature tensile strength and ductility of deposition and casting single crystal superalloys.

| Sample | YS (MPa) | UTS (MPa) | Elongation (%) |
|--------|----------|-----------|----------------|
| A3     | 971.0    | 1068.6    | 16.8           |
| B3     | 1052.6   | 1133.3    | 11.9           |

Figure 9. Histogram of the measured size of \(\gamma'\) precipitates.
4.3. The differences in tensile property between deposition and casting

As seen in table 1, the percent difference of Cr, Co, W, Ti or Ta element is smaller than it of the Al element between A and B. In all composition elements, the boiling point of aluminum is lower than other elements. Therefore, aluminum is the more volatile component in the laser pool or during powder making process. The volatile of aluminum leads to that the volume of $\gamma'$ is lower in deposition than it in casting. Additionally, the size of $\gamma'$ in deposition is bigger than it in casting. It can be concluded that the yield strength and ultimate tensile strength of deposition is smaller than that of casting according to the research of Du [34]. And it agrees with the experimental results in table 5.

As the fracture surface in figure 10, the surface is more smooth in casting than it in deposition. Around the micropores, there are remarkable tearing ridges in the deposition. And the tensile results show that the deposition has more good plasticity. It can be explained by two hands. Firstly, the micropores are bigger and more uneven in casting than it in deposition. Secondly, there are about more than 100 multiples dendrites in the test sample of deposits, and so the deformation in deposition is more uniformly.

Figure 10. Fracture surface of pulling along [001] direction of deposition and casting specimens: (a) A3; (b) B3.
5. Conclusion

(1) A tight control of the process parameters enables to produce of bulk single crystal by laser melting deposition, and the size of the deposition reached $18 \times 10 \times 23$ mm.

(2) The fraction of microscopic holes in deposition is close to it in casting. Owing to the in situ preheating technology, there is no crack in deposition and recrystallization in heat treatment was successfully avoided.

(3) Because the boiling point of aluminum is lower than other alloying elements, aluminum is easily burning loss in the laser pool or during powder making process, and the volume fraction of $\gamma'$-phase is slightly lower in deposition than in it casting.

(4) The mechanical strength of deposition is slightly lower than that of casting sample. The heat treatment needs further research to find a suitable process for the deposition of single crystal superalloy.

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ORCID iDs

Guowei Wang © https://orcid.org/0000-0002-7841-6490

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