DOI: https://doi.org/10.24425/amm.2023.142424

NANFU ZONG[1*], WEIZHAO SUN[1], XINGHONG LIANG[1], TAO JING[1]

ANALYSIS OF TILTED COLUMNAR DENDRITES AT GRAIN BOUNDARIES DURING WIRE AND LASER ADDITIVE MANUFACTURING: A PHASE-FIELD STUDY

Tilted columnar dendritic morphologies are usually existed in wire and laser additive manufactured parts of GH3039 alloy. Overgrowth behaviors induced by the tilted dendritic arrays with a large tilted angle, and the effect of the angle between the growth direction and the direction vertical locally to the solid substrate on primary spacing, solute concentration and morphological evolution have been investigated at both the converging and the diverging grain boundaries through the phase-field simulation. The formation of cracking depends on solidification behaviors including columnar dendrites growth and micro-segregation in the interdendritic region. Furthermore, the effect of the tilted columnar dendrites on the susceptibility of crack is investigated during wire and laser additive manufacturing.

Keywords: Wire and laser additive manufacturing; Tilted columnar dendrites; Overgrowth behavior; Phase field simulation; Grain boundary

1. Introduction

Wire and laser additive manufactured (WLAM) offers advantages as short manufacturing cycle and high flexibility over conventional mechanical manufacture. Since the cooling rate in the molten pool is much faster during additive manufactured processes, the element has not enough time to effectively diffuse. Concentrations of elements are easy to form in the interdendritic region, even micro-segregation in the liquid channel between dendrites, which eventually accelerate the formation of metallurgical defects [1]. When the local stress concentration exceeds the strength limit of the material, the initial cracking would occur.

Tilted columnar dendritic morphologies exist in many additive manufactured processes, and the tilted dendritic arrays are usually with a large tilted angle. The preferred crystallographic orientation plays an essential role on the growth morphologies selection of tilted columnar dendrites [2-4]. Competitive growth occurs between converging grains, which significantly influences the final materials fabricated by additive manufacturing. As to growth direction of columnar dendrites, previous research [5] indicated that, some dendrites can lag behind the other dendrites at the converging grain boundary. Overgrowth at the converging grains was further investigated in directional solidification using phase-field simulations [6-7]. Concentration of solute in the liquid channel between the dendrites could induce a lateral movement at the converging grain boundary, the solute interaction at the grain boundary has a significant impact on the overgrowth of dendrites. Previous experimental investigations [8-9] show that actual growth direction of columnar dendrites undergoes a variation from the thermal gradient direction to the preferred crystallographic orientation with the increase of the pulling velocity. Therefore, competitive growth can also occur between the diverging grains fabricated by additive manufacturing. Mechanical properties of the fabricated materials are related with the tilted growth morphologies and the large columnar dendrites [10]. A few investigations have investigated effects of the crystallographic orientation on overgrowth behaviors at both the converging and the diverging grain boundaries, this investigation can provide theoretical basis to prevent the formation of coarse columnar dendrite in the additive manufactured parts.

In this study, through the phase-field simulations, overgrowth behaviors at both the converging and the diverging grain boundaries have been investigated, and the effects of crystallographic orientation on primary arm spacing, solute concentration and morphological evolution of the columnar dendrite are calculated and explored their internal relationship.

1 TSINGHUA UNIVERSITY, KEY LABORATORY FOR ADVANCED MATERIALS PROCESSING TECHNOLOGY, MINISTRY OF EDUCATION, SCHOOL OF MATERIALS SCIENCE AND ENGINEERING, BEIJING 100084, CHINA

* Corresponding author: zongnan512712_2005@163.com

Analysis the overgrowth behavior of tilted columnar dendrites at grain boundaries can help engineers to control the dendritic growth direction and further improve the product's performance due to major scientific and technological interests.

2. Phase-field modeling and experiments of additive manufacturing

2.1. Quantitative phase field model

In the present study, a multi-scale computational framework that couples finite element method (FEM) and phase field method is developed to simulate the cellular pattern evolution during WLAM. Schematic of the laser molten pool in the macro-scale at early stage is presented in Fig. 1. Detailed derivations and validations of this phase-field model can be found in the references provided above [11-12]. The macroscopic heat transfer model is employed to calculate the transient temperature field and thermal history in the laser molten pool. The local temperature data and molten pool geometry parameters such as melt pool depth a_l and width b_l were extracted from the multi-scale thermal model.

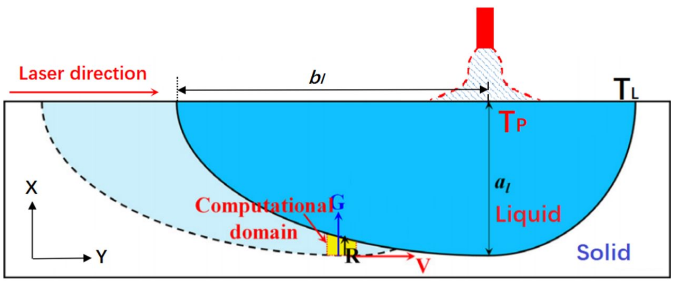

Fig. 1. Schematic of the laser molten pool in the macro-scale at early stage

The pulling velocity ($Vp(t)$) and temperature gradient ($G(t)$) are both the time-dependent. Hence, $Vp(t)$ and $G(t)$ can be expressed as:

$$V_P(t) = \frac{a_l V^2 t}{\sqrt{V^2 t^2 \left(a_l^2 - b_l^2\right) + b_l^4}} \tag{1}$$

$$G(t) = \frac{T_P - T_l}{\sqrt{V^2 t^2 + a_l^2 \dfrac{1 - V^2 t^2}{b_l^2}}} \tag{2}$$

where T_l is the equilibrium liquidus temperature at molten pool edge (K); T_p is the temperature value at the center of molten pool (K); V is the laser scan speed (mm/s); a_l is the melt pool depth (mm); b_l is the melt pool width (mm); t is the time (s); $V_p(t)$ is the pulling velocity (mm/s); $G(t)$ is the temperature gradient (K/mm).

For cubic crystals of GH3039 super alloy, a four-fold anisotropy function is implemented in two-dimensional system to investigate tilted dendrite transition and their growth dynamics during additive manufacturing [2-4]. The selection of crystal growth patterns in solidification has been of fundamental interests in studies of scientific and engineering significance.

2.2. Materials and experimental parameters during WLAM

The technique of WLAM has the characteristics of low cost and high efficiency, which can manufacture the large structural parts of the metal components. The device of WLAM consists of wire feeding device, laser heat source, and temperature measurement system. The chemical composition of the GH3039 super alloy wire is listed in TABLE 1. A single pass-multilayer specimen was built on 45 # steel substrate using the laser processing parameters listed in TABLE 2, the time-dependent solidification parameters of the two-dimensional phase-field model can be determined according to the above parameters.

TABLE 1

Chemical composition of CH3039 super alloy wire

Element	C	Cr	Mo	Al	Ti	Si	Fe	Nb	Ni
Wt%	0.38	19.04	1.86	0.60	0.48	0.19	1.38	0.77	Bal.

TABLE 2

Laser processing parameters

Parameter	Value
Laser power, P (KW)	2
Equivalent laser scan speed, V (mm/s)	4.0
Depth of the laser melt pool, h (mm)	1.8
Width of the laser melt pool, b (mm)	2.6

3. Results and discussion

3.1. Morphological evolution of tilted columnar dendrites

For the columnar dendrites with a large tilted angle, dendrites are patterns with obvious orientational order, tilted dendrite transition and their growth dynamics significantly influences the mechanical properties of the final manufactured materials. Typical microstructure of the specimen has been observed in the longitudinal section and selected Locations. Fig. 2 shows the optical micrographs (OM) of the tilted columnar dendrite produced by WLAM at different selected locations, growth direction of tilted dendritic arrays has a large tilted angle, the growth direction of the columnar dendrite is more sensitive to the crystallographic orientation, especially at the larger pulling velocity and the higher thermal gradient. Magnified tilted dendrite of localized region were observed along the build direction, as shown in Figs. 2(b), 2(c) and 2(d). Some dendrites are deflected and have the remarkable variation of dendrites orientation. In order to explain the columnar dendrites deflection, the morphological evolution and solute concentration of the columnar dendrite has been investigated with and without the crystallographic orientation at about 0.15 s in Fig. 3, the tip positions of all dendrites are almost the same. The tip radius of the columnar dendrite or columnar cell plays an essential role in the oriented morphology formation, which is expressed by the adequate Growth Law [13-14]. According to the mentioned Growth Law the spacing varies significantly with the increasing growth rate. It is well visible that the solid / liquid interface of the dendrites array is planar in the macro-scale. However, the concave / convex shape of the s/l interface is also well visible in the micro-scale. The concave – convex shape (micro-scale) seems to be typical for the eutectic growth [15]. It is evident that the growth (solidification) rate is rapid at the bottom of dendrites (or eutectic phases) and slower at the dendrites tip (or eutectic lamellae tip) forming at the end of the process under investigation. It is well visible that at the bottom of morphology the dendrites (or eutectic lamellae) are narrow. At the threshold growth rate two neighboring dendrites (or two lamellae) transform into one thicker dendrite (or lamella).

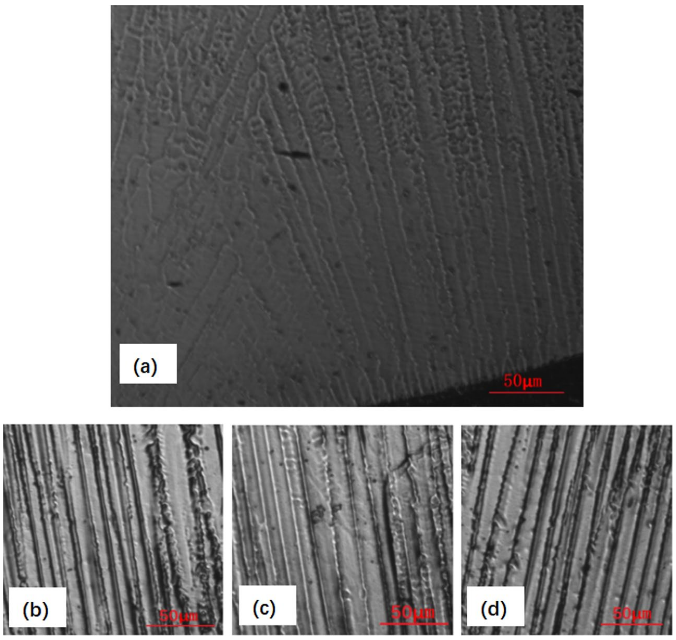

Fig. 2. Optical micrographs of the tilted columnar dendrite produced by WLAM at different selected locations: (a) low magnification micrograph of dendrite; (b) typical tilted dendrite along left side; (c) conventional dendrite; (d) typical tilted dendrite along right side

The behavior of the green dendrites (or lamellae in the case of the eutectic growth). Moreover, the green dendrites become green / blue ones. It means that growth rate decreases during solidification, and consequentially an intensity of the Cr – solute micro-segregation (as observed across a given dendrite (or lamella)) is subjected to some changes due to both: the possible appearance of the diffusion into the solid (back-diffusion) and evolution of the partition ratio with the growth rate [16]. Analogously, the inter-dendritic region (or other eutectic phase) changes its color from yellow into red one. The inter-dendritic region (eutectic phase) presents the concave shape of its s/l interface. In the case of the eutectic growth a transition layer just between the liquid and lamellae of the solid can be expected [17].

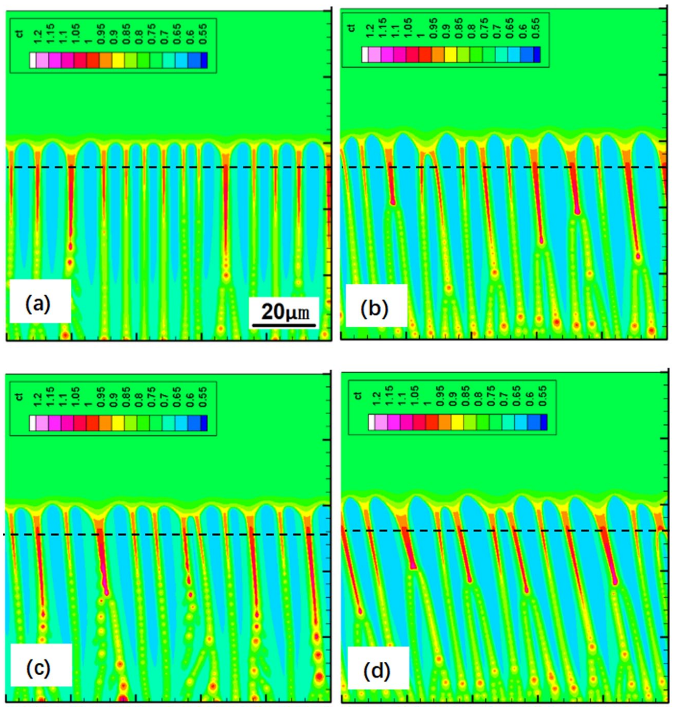

Fig. 3. The morphological evolution and Cr – solute concentration of the columnar dendrite at transient condition of 0.15 s: (a) crystallographic orientation 0°; (b) crystallographic orientation 15°; (c) crystallographic orientation 30°; (d) crystallographic orientation 45°

The suggested threshold growth rate which can be distinguished in Fig. 3, in fact, can be treated as the rate which divides the performed solidification into rapid solidification and slower one. In the case of eutectic growth, the inhomogeneity of the liquid ahead is always formed. This inhomogeneity can be described due to the solution to the diffusion equation as shown for a slow solidification [18] and for a rapid solidification [19]. It is evident that the protrusion of the green-blue phase (dendrite or eutectic lamella-the leading phase) over the red one (inter-dendritic region or second eutectic lamella-wetting and delayed phase) is significant (micro-scale) [16]. The fluctuation of the solute concentration is the major reason for the deflection of columnar dendrites in an alloy system [8-9]. As the morphological evolution shown in Fig. 3, it can be predicted that the deflection angle of columnar dendrites increases slightly with the increase in crystallographic orientation, and the columnar dendritic agrees well with the experimental results as shown in Fig. 2. The local line refer to Fig. 3 has been taken to calculate the Cr–solute concentration. Solute concentrations in Fig. 3 are enlarged and compared under different crystallographic orientation as shown in Fig. 4.

Cr – solute concentration at selected locations increases with the increasing of crystallographic orientation from Fig. 3(a) to Fig. 3(d). Since the solidification rate in the molten pool

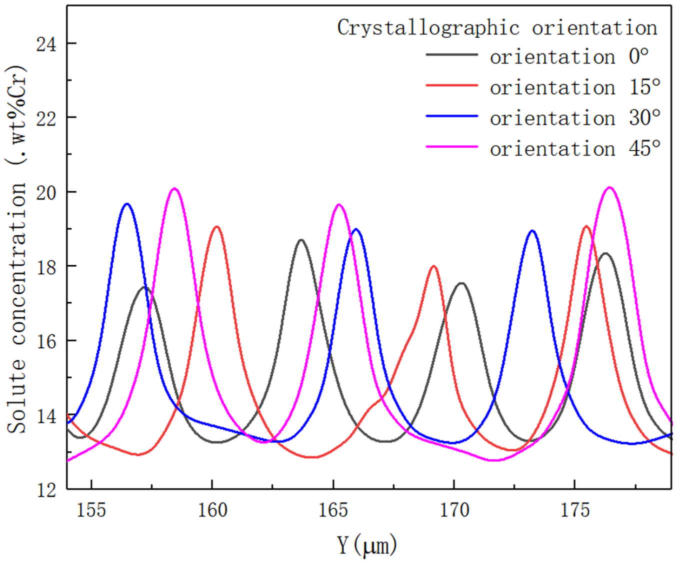

Fig. 4. Local lines of Cr – solute concentration under different crystallographic orientation

is much faster than the general solidification behaviors, the element does not have enough time to diffuse and concentration of Cr is extracted to the liquid channel between dendrites increases. In addition, the enrichment of micro-segregation can enlarge the primary dendrites, while the lateral branches grow strongly and some branches have become thick dendrites as shown in Fig. 5.

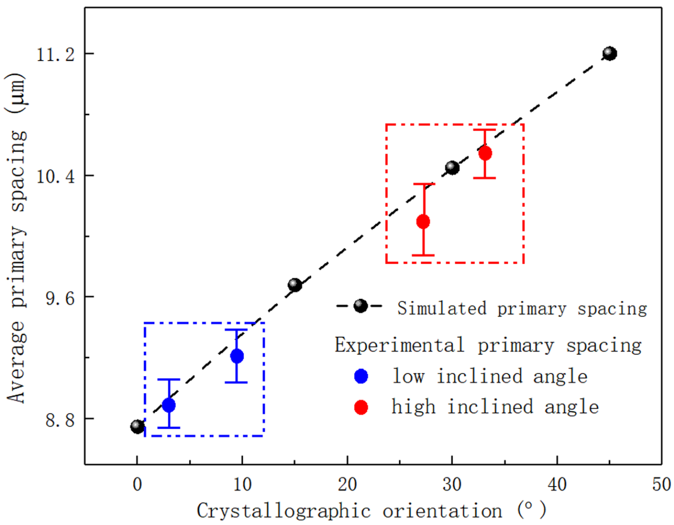

Fig. 5. Simulated and experimental average primary spacing

The thin interface analysis used for a quantitative phase-field model has been taken to simulate the dendrites evolution. The theoretical equation developed for columnar dendrites (or columnar cells) worked out with the application of the marginal stability criterion [16]. To understand the effect of crystallographic orientation on primary spacing, we systematically changed crystallographic orientation from 0° to 45°. When crystallographic orientation is large, average primary spacing largely fluctuates. The measured average primary spacing of experimental dendritic morphologies gives better agreements with simulation results. The primary spacing is one of the important factors characterizing the columnar dendrite structure, this experimental average primary spacing value increases with an increase in the deflection of columnar dendrites, and its dependence is consistent with the theoretical models of crystallographic orientation.

3.2. Overgrowth of tilted columnar dendrite at the grain boundaries

Overgrowth behaviors of columnar dendrite can enlarge the primary arm spacing at the grain boundaries. The tilted growth of columnar dendrites plays an important role in the growth pattern of Cr – solute concentration and morphological evolution, two types of overgrowth behaviors at both the converging and the diverging grain boundaries have been investigated, and the effects of crystallographic orientation on primary arm spacing, Cr – solute concentration and morphological evolution of the columnar dendrite at transient condition.

TABLE 3

Configurations of columnar dendrite orientations in phase-field simulations.

Cases	Type	θ_L (°)	θ_R (°)
diverging case	type I	–30	**10**
converging case	type II	10	**–30**

Configurations of columnar dendrite orientations in phase-field simulations are presented in TABLE 3. θ_L and θ_R dendrites were the inclination angles of the left and right grains at the grain boundaries to the thermal gradient direction, respectively.

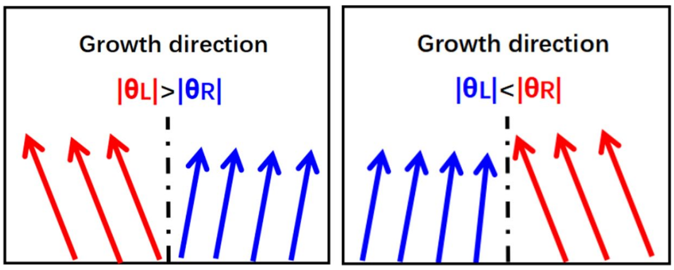

Fig. 6. A schematic illustration of columnar dendrite growth at the diverging and converging grain boundaries

A schematic illustration of columnar dendrite growth at the diverging and converging grain boundaries is shown in Fig. 6, every single arrow indicates a columnar dendrite, which is shown as red and blue, respectively, and the black dashed lines denote the trajectories of the grain boundaries.

Cr – solute concentration and morphological evolution of Type I columnar dendrite under different times are shown in Fig. 7. The columnar dendrite at the diverging grain boundaries

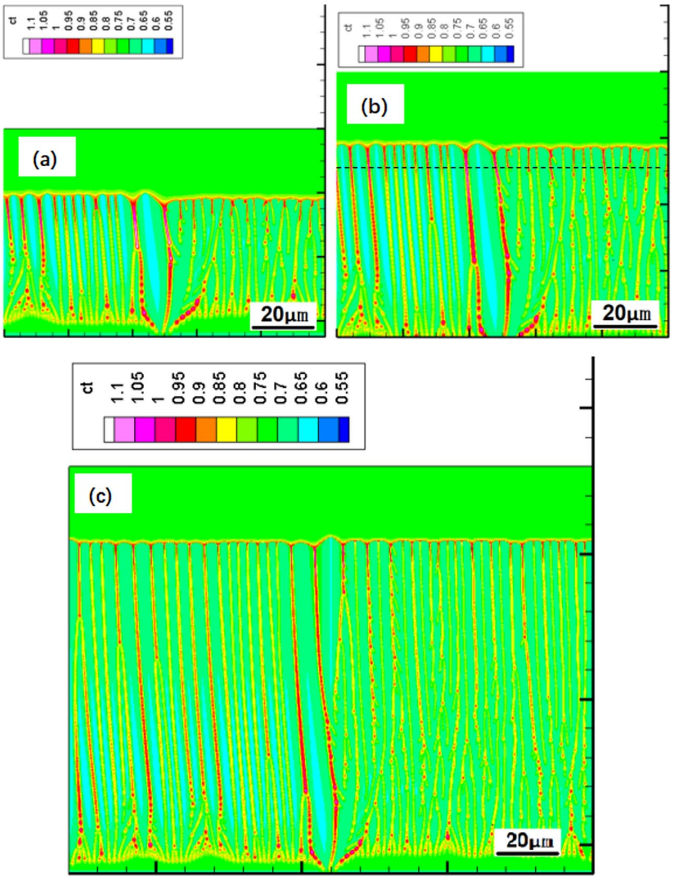

Fig. 7. Cr – solute concentration and morphological evolution of Type I columnar dendrite under different times: (a) 0.19 s; (b) 0.23 s; (c) 0.29s

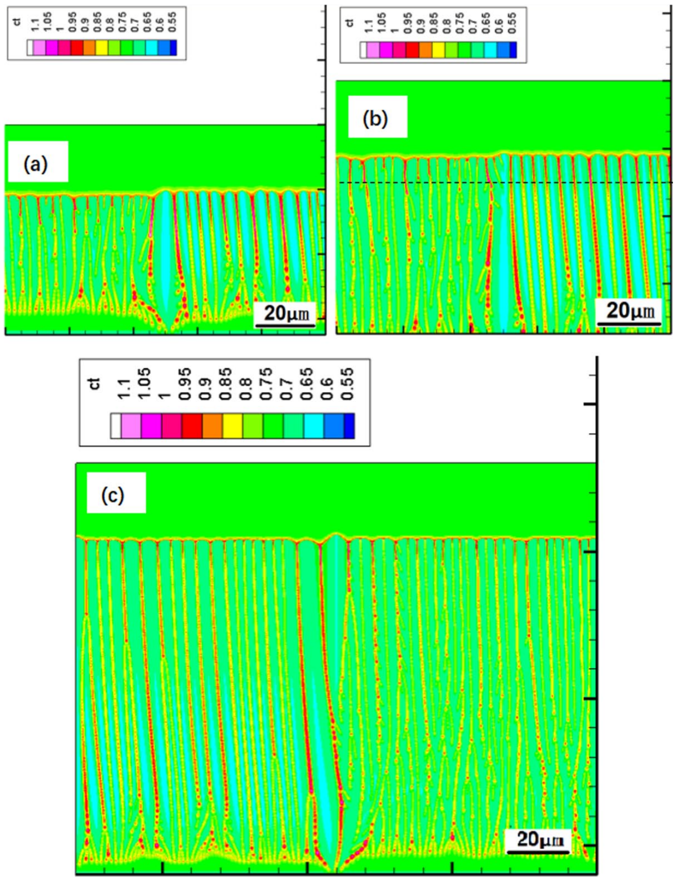

Fig. 8. Cr – solute concentration and morphological evolution of Type II columnar dendrite under different times: (a) 0.19 s; (b) 0.23 s; (c) 0.29s

showed a large variation, which were related to overgrowth of tilted columnar dendrite. The columnar dendrites at the diverging grain boundaries have different inclination direction. When the columnar dendrites are inclined in opposite lateral direction as type I in Fig. 7, the maximum columnar dendrite spacing at the grain boundary was larger than that inside the dendrites. The left part columnar dendrite changed from the trajectories of the grain boundaries to the left columnar dendrite side, the right part columnar dendrite changed from the trajectories of the grain boundaries to the right columnar dendrite side. Furthermore, overgrowth behavior of columnar dendrite can branch new dendrites at diverging grain boundaries with local fragments, overgrowth behavior can occur more easily when there was a difference between the inclination angles of two adjacent grains.

Cr – solute concentration and morphological evolution of Type II columnar dendrite under different times are shown in Fig. 8. The overgrowth of columnar dendrites could also occur when there was a difference between the inclination angles of two converging grains, the simulation results indicated that the dendrite spacing was adjusted to a stable range by the elimination of existing unstable dendrites. It is well visible in both Fig. 7 and Fig. 8 that one dendrite is significantly larger among others dendrites. It can be concluded that this is a beginning of the phenomenon named as the solitary wave. The solitary wave begins still at the planar s/l interface and usually leads to the formation of non-planar s/l interface [11-12]. The maximum columnar dendrite spacing at the converging grain boundary was greater than that of tilted columnar dendrites. Local lines of Cr – solute concentration under various cases are shown in Fig. 9. The dashed lines are represented the upper and lower limits of the Cr – solute concentration boundary, and the local lines are located at the dashed line in Fig. 7(b) and Fig. 8(b). The upper limit of Cr – solute concentration with crystallographic orientation 30°(–30°) is more larger than that with crystallographic orientation 10°(–10°), however, the lower limit of Cr–solute concentration with crystallographic orientation 30°(–30°) is smaller than that without crystallographic orientation 10°(–10°).

Compared with the directional solidification, the columnar dendrite morphology transfers from symmetrical dendrite to tilted asymmetrical dendrite and the lateral growth was more severe. Moreover, increasing the deflection angle can lead to uneven composition of material matrix, especially at the converging grain boundaries.

Fig. 10 shows the optical micrographs showing the solidification cracking during wire and laser additive manufacturing. The cracks are always located at the grain boundaries between different inclination angles of columnar dendrites. The formation of cracking depends on solidification behaviors including columnar dendrites growth and micro-segregation in the interdendritic region. When the local stress concentration exceeds the

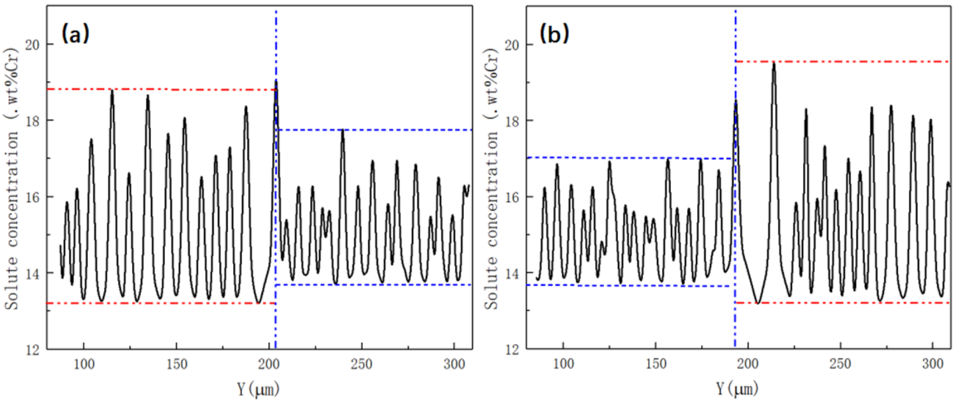

Fig. 9. Local lines of Cr – solute concentration under various cases: (a) type I; (b) type II

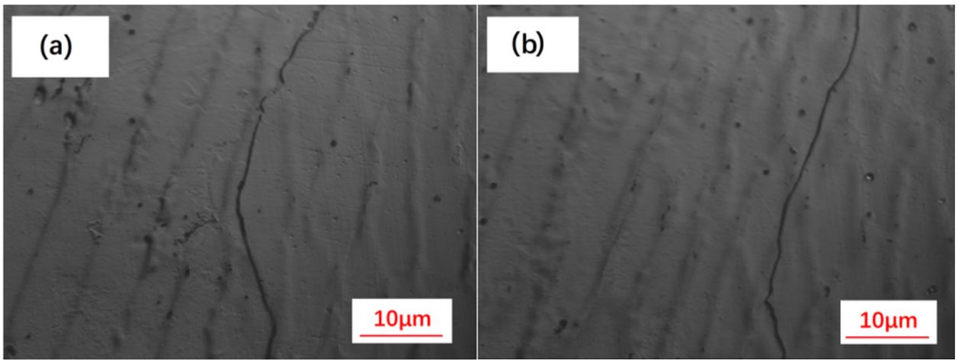

Fig. 10. Optical micrographs showing the solidification cracking during wire and laser additive manufacturing

strength limit of the material, the cracking would occur. Since the solidification rate is much faster during the additive manufacturing, the elements are very easy to concentrate in liquid channel between dendrites and the susceptibility of crack is very high. As shown in Fig. 7 and Fig. 8, increasing of inclination angles of the columnar dendrite makes the average columnar dendrite spacing increase, especially at the grain boundaries. Therefore, overgrowth and micro-segregation are easily generated at both the converging and the diverging grain boundaries of tilted columnar dendrites, which may lead to the appearance of cracking in the additive manufactured parts.

4. Conclusions

A two-dimensional phase-field simulation has been developed to investigate the growth competition between the grain interior and the grain boundaries in the molten pool of GH3039 nickel-based alloy, with different angle between the growth direction and the direction vertical locally to the solid substrate. The major conclusions in this paper are summarized as follows:

(1) A cluster columnar dendrites are mainly distributed at the bottom of the cladding layers, tilted columnar dendritic morphologies are usually existed in wire and laser additive manufactured parts of GH3039 alloy, the other typical columnar dendrites have the angle between the normal direction of substrate surface.

(2) The deflection angle of columnar dendrites increases slightly with the increase in crystallographic orientation. The experimental average primary spacing value increases with an increase in the deflection of columnar dendrites.

(3) Overgrowth behaviors were investigated both the converging and the diverging grain boundaries through two-dimensional phase-field simulation, and the effect of dendrite orientation on relative overgrowth behavior in the lateral direction were closely analyzed. The columnar dendrite at the grain boundaries showed a large variation, the maximum columnar dendrite spacing at the grain boundary was larger than that inside the dendrites. Moreover, increasing the deflection angle can lead to uneven composition of material matrix, especially at the converging grain boundaries in the wire and laser additive manufacturing parts.

(4) The cracks are always located at the grain boundaries between different inclination angles of columnar dendrites in the additive manufactured parts. Overgrowth and micro-segregation are easily generated at both the converging and the diverging grain boundaries of tilted columnar dendrites, which may lead to the appearance of cracking.

Acknowledgments

The present work is financially supported by The National Key Research and Development Program of China No. 2017YFB1103700.

REFERENCES

[1] T. Guraya, S. Singamneni, Z.W. Chen, Microstructure formed during selective laser melting of IN738LC in keyhole mode, Journal of Alloys and Compounds **792**, 151-160 (2019).

[2] T. Ihle, Competition between kinetic and surface tension anisotropy in dendritic growth, The European Physical Journal B **16**, 337-344 (2000).

[3] E. Ben-Jacob, P. Garik, The formation of patterns in non-equilibrium growth, Nature **343**, 523-530(1990).

[4] Z. Wang, J. Li, J. Wang, Predicting growth direction of tilted dendritic arrays during directional solidification, Journal of Crystal Growth **328**, 108-113 (2011).

[5] Y.Z. Zhou, A. Volek, N. Green, Mechanism of competitive grain growth in directional solidification of a nickel-base superalloy, Acta Materialia 56, 2631-2637 (2008).

[6] H. Yu, J. Li, X. Lin, L. Wang, W. Huang, Anomalous overgrowth of converging dendrites during directional solidification, Journal of Crystal Growth **402**, 210-214 (2014).

[7] J. Li, Z. Wang, Y. Wang, J. Wang, Phase-field study of competitive dendritic growth of converging grains during directional solidification, Acta Materialia **60**, 1478-1493 (2012).

[8] C. Gandin, M. Eshelmann, R. Trivedi, Orientation dependence of primary dendrite spacing, Metallurgical and Materials Transactions A 27A, 2727-2739 (1996).

[9] T. Haxihimali, A. Karma, F. Gonzales, M. Rappaz, Orientation selection in dendritic evolution, Nature Materials **5**, 660-664 (2006).

[10] M. Cloots, P.J. Uggowitzer, K. Wegener, Investigations on the microstructure and crack formation of IN738LC samples processed by selective laser melting using Gaussian and doughnut profiles, Materials & Design **89**, 770-784 (2016).

[11] Z. Wang, S. Ma, W. Sun, M. Zhang, T. Jing, H. Dong, Cellular tip splitting instability during transient growth, Computational Materials Science **155**, 364-372 (2018).

[12] W. Zheng, Z. Dong, Y. Wei, K. Song, Onset of the initial instability during the solidification of welding pool of aluminum alloy under transient conditions, Journal of Crystal Growth **402**, 203-209 (2014).

[13] P. Kwapisiński, Z. Lipnicki, A.A. Ivanova, W. Wołczyński, Role of the Structural and Thermal Peclet Numbers in the Brass Continuous Casting, Archives of Foundry Engineering **17** (2), 49-54 (2017).

[14] W. Wołczyński, Entropy Production in the Stationary Eutectic Growth, Archives of Metallurgy and Materials **65** (1), 403-416 (2020).

[15] W. Wołczyński, Formation of Concave-Convex Interface Shape during Oriented Eutectic Growth, Crystal Research and Technology **26**, 173-178 (1991).

[16] W. Wołczyński, Pattern Selection in the Eutectic Growth-Thermodynamic Interpretation, Archives of Metallurgy and Materials **65** (2), 653-666 (2020).

[17] W. Wołczyński, Contribution to Transition Layer Determination for Oriented Eutectic Growth, Crystal Research and Technology **25**, 1433-1437 (1990).

[18] W. Wołczyński, Concentration Micro-Field for Lamellar Eutectic Growth, Defect and Diffusion Forum **272**, 123-138 (2007).

[19] W. Wołczyński, Development of the Jackson and Hunt Theory for Rapid Eutectic Growth, Archives of Metallurgy and Materials **63** (1), 65-72 (2018).