Investigation of growth of single crystal SRR99 superalloy under microgravity using 50-meter-high drop tube

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Abstract. The SRR99 Ni-base single crystal superalloy samples were partially remelted and then solidified during free falling in a 50-meter-high drop tube. For comparison, samples solidified statically, i.e., in unit gravity, were also conducted under equivalent condition. Under the both conditions there were areas where columnar dendrites epitaxially grew from the seed crystals. The epitaxial growth of columnar dendrites in the samples solidified during free fall was subject to the influence of microgravity. It was found that the primary dendrite arm spacing of the sample solidified under microgravity was apparently larger than that of the sample solidified under gravity. The secondary arm lengths were also longer in the microgravity sample. Besides, the distributions of heavy elements such as W and Ta along the core of a primary dendrite arm were found a little bit different between the samples solidified under the two conditions. Based on the above experimental results, the effects of gravity on solidification microstructure and solute distribution were analyzed.

1. Introduction
Nickel-base single crystal (SX) superalloys have been widely applied in advanced gas turbines and jet engines for their superior strength and fracture resistance at elevated temperatures [1, 2]. In order to obtain a combination of good mechanical strength and superior high temperature capabilities, the SX superalloys have been heavily alloyed with elements of low densities (Al, Ti) as well as elements of high densities (Ta, W, Re). In particular, the addition of these refractory elements has contributed greatly to the improvement in high-temperature performance of SX superalloys [3-5].

As the rising levels of refractory alloying additions, grain-defect formation during directional solidification of SX superalloys has become an increasingly important problem. Although the formation of these defects is resultant of a number of factors, convective instability induced by gravity during directional solidification is regarded as the precursor event [6]. Consequently, understanding the effects of convection is of great importance on the solidification microstructures of SX superalloys. However, due to the complicated nature of convection and the sophisticated interaction between fluid flow and microstructure evolution, the influence of gravity-induced convection has not been recognized thoroughly.

Owing to the absence of gravity and consequently the related phenomena, the utilization of microgravity environment is of great importance to study the effect of gravity on solidification [7, 8].

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Among all the microgravity facilities, long drop tubes are powerful instruments in earthbound laboratories [9-11]. Therefore, in the present paper, the effect of gravity on the microstructures of a SX superalloy was investigated by using a long drop tube.

2. Experimental
The SRR99 single crystal superalloy used for the solidification experiment was prepared by the Bridgman technique. The nominal composition of this alloy is listed in table 1. The microgravity experiments were carried out in a 50 m long drop tube. Cylindrical samples of 6 mm in diameter and 20 mm in height were enveloped in high purity corundum crucibles. When doing experiment, the drop tube was evacuated to $6 \times 10^{-3}$ Pa. Then, the top part of the sample was partially heated by a spiral induction heater installed at the top of the drop tube. The temperature of the top surface of the sample was monitored using an infrared pyrometer. The schematic diagram of the heating apparatus in the drop tube is shown in figure 1. When the upper part of the sample was remelted and the lower part was still solid, the power of the spiral induction coil was turned off. At the same time, the sample together with the crucible was released and dropped down, subject to a microgravity condition of about $10^{-6}$ $g_0$ ($g_0$ is normal gravity) for about 3.24 s. At last, the sample was quenched into silicone oil when landing at the bottom of the drop tube. In the present work, the heating time was about 13 s. Such kind of the sample was referred to as $\mu$g sample hereinafter. For comparison, the same remelting and solidification experiments were carried out, except for the 50 m free falling. The sample obtained in this case was referred to as 1g sample hereinafter. The re-solidified parts of all the samples were cut both longitudinally and transversely. The microstructures on the sections were observed under optical microscopy after mounting, grinding, polishing and etching the samples. Primary dendrite arm spacings were measured on the transverse sections at almost the same distance from the initial remelting interface. Besides, the lengths of the secondary dendrite arms were also measured on the cross sections, since on the cross sections the four secondary dendrite arms growing from one primary dendrite arm can be observed on the same plane. Electron probe microanalysis (EPMA) was applied to measure the element distributions in the primary dendrite arms on the longitudinal sections of the samples. For both types of samples, the primary dendrite arms measured were chosen to be almost at the same place on longitudinal sections. Samples were lightly etched and point analyzes were performed about every 200 $\mu$m along the dendrite core from the remelting interface to its tip. During X-ray sampling, a spot size of 1 $\mu$m was used with an accelerating voltage of 15 kV and a beam current of 100 nA. All the elements were analyzed at each point utilizing pure elemental or mineral standards. Then, the raw data were processed by a standard ZAF correction program to give normalized weight percentages of each element.

| Element | Al  | Ti  | Cr  | Ta  | W   | Co  | Ni  |
|---------|-----|-----|-----|-----|-----|-----|-----|
| Weight (%) | 5.47 | 2.14 | 8.39 | 2.92 | 9.47 | 5.01 | Bal |

3. Results and Discussion
The remelted length is about 6 mm for both $\mu$g sample and 1g sample. Figure 2 illustrates the thermal histories of the two types of the samples. It shows that both samples were heated rapidly at the initial 6 s, which could ensure the samples to be remelted partially. Then, the temperatures increased very slowly for about 7 s, which should correspond to the melting processes of the samples. During the entire period of induction heating for 13 s the temperature characteristics of both $\mu$g sample and 1g sample were almost the same, indicating that the initial solidification conditions for both samples are almost the same. For 1g sample, as soon as the power was turned off, the temperature of the top surface of the sample kept almost constant for less than 4 s. Then, the temperature abruptly increased about 47 °C, and soon after it decreased monotonously. Since the solid part of the sample could act as
a seed crystal, the liquid part of the sample began to solidify immediately once the heating process stopped. In other words, the samples started to solidify at the moment of the 13th s. During the solidification, although the temperature at the solid-liquid interface was cooling down, because the top end of the sample was the final site to solidify, it showed there a plateau on the temperature profile of 1g sample due to the balance between the heat loss and the release of latent heat. The rapid increase of temperature after the plateau may be due to the recalescence phenomenon at the last stage of solidification. Therefore, it is reasonable to deduce from the temperature profile that 1g sample should completely solidify in 4 s. It should be pointed out that the temperature of μg sample could not be monitored after it was released to fall down in the drop tube. However, owing to the similar cooling conditions for both types of the samples, μg sample should experience a nearly identical thermal history and solidify almost totally during the 3.24 s of free fall.

The optical micrographs of figure 3 show the microstructures on longitudinal sections of μg sample and 1g sample, respectively. The coarse dendrites at the lower part are the microstructures of the seed crystals. At the remelting interface, straight columnar dendrites grew epitaxially from the original microstructures. Besides, at the surfaces of the cylindrical samples, some wild dendrites nucleated from the crucible surfaces and grew toward the inside of the samples, interfering and finally blocking the epitaxial growth of the dendrites from the seeds. Comparatively, the maximum length of the epitaxially grown columnar dendrites in 1g sample is about 100 μm longer than that in μg sample. It seems that the macrostructures of both samples show little difference. Since the columnar dendrites in the vicinity of the initial growth interface grew under an approximate directional solidification condition, and particularly, this part of microstructure formed undoubtedly under microgravity for μg sample, in the following the detailed features of these columnar dendrites are investigated.

The transverse microstructures of μg sample and 1g sample are shown in figure 4. It is noticeable that the dendritic trunks in μg sample are coarser than those in 1g sample. For instance, secondary dendrite arms are longer in μg sample. In addition, it is clearly seen that extensive tertiary dendrites grew from secondary branches in μg sample, whereas there are only a few tertiary dendrites observed in 1g sample, as the white arrows in figure 4 indicate. Figure 5 shows the distribution of secondary
dendrite arm lengths. For both types of the samples, the minimum secondary dendrite arm lengths are the same. However, the maximum length in µg sample is 20 µm longer than 1g sample. Moreover, the distribution of secondary dendrite arm lengths is more scattered in µg sample. The discrepancy between the maximum and the minimum frequencies is about 20%. On the contrary, in 1g sample the distribution is more centralized, mainly concentrating in the range from 20 µm to 80 µm. The difference between the maximum and the minimum frequencies is about 37%. Another interesting phenomenon is that a great number of fine primary dendrites exist in 1g sample, while few such dendrites can be found in µg sample, as the black arrow in figure 4 (b) indicates. Figure 6 demonstrates the distribution of primary dendrite arm spacing corresponding to figure 4, the dashed lines are Gaussian fits through the data points. Although the range of primary arm spacing varying from 40 µm to 140 µm is unaltered for both types of samples, the distribution of primary arm spacing is more symmetrical in µg sample. Besides, at the peak of the Gaussian curve, the primary arm spacing is 71.5 and 58.7 µm for µg sample and 1g sample, respectively. The average primary arm spacing of µg sample and 1g sample is about 85 and 71 µm, respectively.

Figure 4  Typical microstructures of SRR99 alloy on transverse sections solidified under microgravity (a) and normal gravity (b).

Figure 5  Distribution of secondary arm lengths on cross section of SRR99 alloys solidified under microgravity (a) and normal gravity (b).

Figure 7 illustrates the solute concentrations measured along the core of a typical epitaxially grown dendrite. It is clearly that, except for W and Ta, there are very little variations in solute compositions along the dendrite core with the distance up to 1.4 mm from the remelting interface for both types of samples. As alloy solidification proceeds, the composition of W in µg sample increases gradually from about 9% to 10%. In 1g sample however, the composition of W fluctuates at the first stage of solidification and then keeps almost constant with the increase of solidification distance. For the element Ta, it is nearly unchanged within the first half length of the columnar dendrite in µg sample, and then it oscillates with the increase of distance. While in 1g sample, the concentration of Ta decreases with the increase of the distance from the remelting interface.

The primary dendrite arm spacing in µg sample is much larger than that in 1g sample, suggesting that gravity has a significant effect on the growth of dendritic microstructures. Under microgravity
conditions, the gravity driven convection becomes weak, which will decrease the mass and heat transport in the liquid phase [12]. Therefore, the growth rate of columnar dendrites will be decelerated. This is in accordance with the shorter columnar dendrites observed on the longitudinal section for μg sample. According to the model of dendrite growth, lower growth rate leads to larger primary dendrite arms spacing [13]. Resultantly, due to the decrease of growth rate, the primary dendrite spacing in μg sample solidified under microgravity increases. The increase of primary dendrite spacing provides larger spaces for the growth of secondary dendrite arms, which thus grow longer along the direction perpendicular to the primary dendrite trunks. Inversely, due to the enhanced fluid flow and consequently rapid growth rate, the primary dendrite spacing decreases in 1g sample. The decrease of primary dendrite arm spacing is characterized by the growth of tertiary dendrite arms, which are promoted by the gravity driven convection and become new primary dendrite arms. Due to the limitation of existing primary dendrites in mushy zone, the tertiary dendrite arms turn out to become fine primary dendrite arms.

Figure 6  Distribution of primary arm lengths on transverse section of SRR99 alloys solidified under microgravity (a) and normal gravity (b).

Figure 7  Solute element concentrations plotted vs. distance from the remelting interface along the core of a primary dendrite arm for SRR99 alloy solidified under microgravity and normal gravity.

The solute distributions along the core of a primary dendrite arm show little difference for the elements of Al, Cr, Co and Ti. However, for the heavy elements such as W and Ta, the concentrations present different features as the growth of primary dendrites proceeds. According to the study by
D'Souza et al [1], the intrinsic measurement uncertainties of the elemental concentrations are beyond an order of magnitude smaller than the scatters. Therefore, the variations in elemental compositions of W and Ta in figure 7 should be caused by the factor of gravity, i.e., gravity may affect the distribution of higher density elements during alloy solidification, even though it shows little effects on the relative lighter elements. Since the solute concentrations along the core of the primary dendrite depend on the solute boundaries at the dendrite tip, gravity may affect the solute distribution of W and Ta by means of changing the solute boundaries. The different behaviors of W and Ta elements in both types of samples should attribute to the different partition coefficients, which are larger than 1 for W and smaller than 1 for Ta. In order to confirm these assumptions, further investigations should be done.

4. Conclusions
Solidification of SRR99 Ni-base single crystal alloy under microgravity and normal gravity conditions was carried out by using a 50 m long drop tube. Samples solidified under microgravity are characterized by coarser dendritic trunks, a little bit longer secondary dendrite arms and scattered distribution of secondary dendrite arm lengths, while samples solidified in normal gravity show shorter secondary dendrite arms and more concentrated arm length distribution. There are a great number of fine dendrites in gravity samples, whereas microgravity samples have mainly coarse dendrites. The average primary arm spacing is about 85 μm for microgravity sample, while it is about 71 μm for gravity sample. Gravity has insignificant influence on the solute distribution for the elements of Al, Cr, Co and Ti in the present work. However, for the heavy elements such as W and Ta, the solute distributions during solidification seem to be sensitive to gravity.

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