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# Primary and secondary dendrite spacing of Ni-based superalloy single crystals

SLOBODANKA KOSTIĆ[1], ALEKSANDAR GOLUBOVIĆ[1*#] and ANDREJA VALČIĆ[2]

[1]*Institute of Physics, Pregrevica 118, P.O. Box 68, 11080 Zemun and* [2]*Faculty of Technology and Metallurgy, Karnegieva 4, 11000 Belgrade, Serbia*

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*Abstract*: Ni-based superalloy single crystals were grown by different methods (gradient method and Bridgman technique with spontaneous nucleation and with seed). In all crystal growth experiments using the Bridgman technique, the temperature gradient along the vertical furnace axes was constant ($G$ = 33.5 °C/cm). The obtained single crystals were cut, mechanical and chemical polished, and chemically etched. Using a metallographic microscope, the spacing of the primary and secondary dendrites was investigated. The dendrite arm spacing (DAS) was determined using a Quantimet 500 MC. The obtained results are discussed and compared with published data.

*Keywords*: superalloy; crystal growth; single crystal; dendrites; quantimeter.

## INTRODUCTION

Ni-based, single crystal alloys have applications in advanced gas turbines and jet engines, due to their improved mechanical properties, creep resistance, thermal fatigue resistance and high melting point as compared to the conventionally prepared Ni-based superalloys. The use of these expensive materials as gas turbine components requires stability of the microstructure as well as of the chemical composition. During fabrication and repair of the components, microstructural changes may occur, which can affect the mechanical properties. The main crystallization parameters having a considerable influence on the microstructure and physical properties of the material are the temperature gradient ($G$), the crystal growth rate ($R$), and the concentration of the alloying elements ($c_0$).[1]

Ni-based superalloys are biphasic compounds, consisting of a face-centered cubic γ matrix and γ' precipitates. Dendrites are the most common growth morphology during the solidification of metals, alloys and other substances.[2] Ni-based superalloys single crystals also have a dendritic structure. Due to the need

* Corresponding author. E-mail: golubovic@phy.bg.ac.rs

# Serbian Chemical Society member.

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to predict and control the microstructure of cast alloys, dendritic growth has been extensively studied. Since metallic alloys are opaque and have a high melting point, which impedes precise control of the solidification condition, direct observation of growing dendrites in transparent analogues has become a powerful tool for investigating microstructure formation under controlled conditions.[3]

A number of studies have indicated the effect of microstructure and, particularly, of dendrite spacing upon mechanical properties.[4] Dendritic structures can be more important in the prediction of mechanical properties than grain size. The improved mechanical characteristics of cast structures having smaller dendrite spacings are largely due to the shorter wavelength of the periodicity of the microsegregation. Much of the initial research devoted to dendritic coarsening was focused on the measurement of the secondary dendritic arm spacing, $\lambda_2$, and the proposal of a mechanism aimed at describing the evolution of this secondary arm spacing.[5] The aim of this study was to investigate the properties of the primary and secondary dendrites of Ni-based superalloy single crystals in order to improve the application of the superalloy.

## EXPERIMENTAL

The composition of the superalloy 444 (Ed Fagan Inc., USA) was (in mass %): 8.6 % Cr, 1.98 % Ti, 5.1 % Al, 11.1 % W, 0.91 % Nb and 72.31 % Ni.

Single crystals of the Ni-based superalloy were grown by the Bridgman (spontaneous nucleation or from a polycrystalline seed) technique[6,7] and the gradient method.[6]

The temperature gradient along the vertical furnace axes for the Bridgman technique was constant ($G$ = 33.5 °C/cm).

All the samples were longitudinally cut, then mechanically and chemically polished, and finally selectively etched.[7]

All photographs were obtained using a metallographic microscope (PM10-35ACS, Olympus, Japan). The magnifications were 20.

Using an automatic device for quantitative picture analysis, Quantimet 500 MC (Leica Cambridge Ltd., Cambridge, UK), and the linear measuring method applied to the samples of superalloy, the dendrite arm spacing (*DAS*) were determined.

## RESULTS AND DISCUSSION

The process of single crystal growth by the gradient method was performed in the standard way.[6] The seed used to commence the crystallization was a fine--grained material of the same alloy. The experiments were performed at different cooling rates ($V$), which remained almost constant during the experiment. The best result was obtained with a cooling rate of 3.4 mm/min. Polycrystalline structure was approximately 10 mm from the top of the seed, and after that it was a single crystal to almost the end of the sample. Although the temperature gradient along the vertical furnace axes was not constant, the growing rate increased during the experiment by a factor of approximately 10.

The crystal growth by the Bridgman technique was performed in two different ways: a) the growth commenced from a completely molten alloy (spontaneous nucleation) or from a polycrystalline seed, formed by a relatively slow nucleation and b) the growth started from a polycrystalline seed, which had a more coarse structure than the seed used in the gradient method.

The temperature gradient along the vertical furnace axes was fixed and the same for both types of Bridgman techniques ($G$ = 33.5 °C/cm). The crystal growth rate was changed from 1 mm/min to 11 mm/min in order to obtain the best value. It was found in the literature that other authors[4] worked in a similar way, but fixed the crystal growth rate while changing the temperature gradient. In addition, these authors had a transparent material, while in this study a real alloy was employed.

A microphotograph of the sample grown by the Bridgman technique with spontaneous nucleation, with the crystal growth rate of 2 mm/min, is shown in Fig. 1. Samples grown by Bridgman technique with seed were grown with uniform rate or with different rates (the rate was changed in the process of crystal growth), in order to investigate the properties of dendrite spacing. The microphotographs of the same crystal grown by the Bridgman technique with seed at growth rates of 6 mm/min and 2 mm/min are shown in Figs. 2 and 3, respectively.

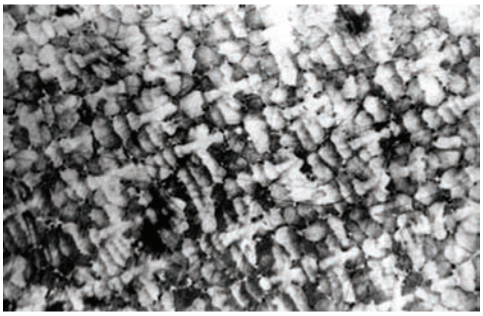


Fig. 1. Photograph showing the primary dendrite spacing in a Ni-based superalloy single crystal obtained by the Bridgman technique with spontaneous nucleation. The crystal growth rate was 2 mm/min and the length of the single crystal was 11.5 cm. The photograph was taken at 12–15 mm from the beginning of the crystal with a magnification of ×24.

Fig. 2. Photograph showing the primary dendrite spacing in a Ni-based superalloy single crystal obtained by the Bridgman technique with seed. The crystal growth rate was 6 mm/min and the length of the single crystal was 6.36 cm. The photograph was taken at 2 cm from the seed with a magnification of ×24.

It is clear that all the samples were single crystals and the various crystal growth methods can serve as a tool for a better understanding of the processes into non-transparent materials. For Ni-based superalloys, a single crystal with a dendritic structure is the common type of structure, as the process of thermal homogenization always follows the process of crystal growth.[8] Crystal growth was in the [001] direction of the all samples, which is in accordance with literature data.[3]

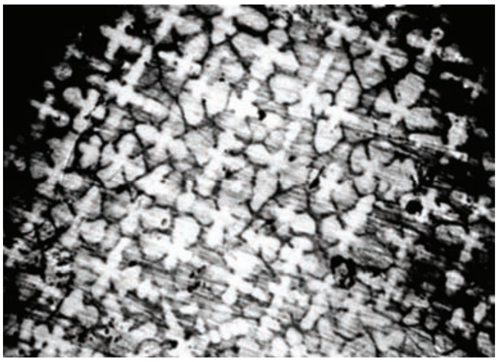


Fig. 3. Photograph showing the primary dendrite spacing in a Ni-based superalloy single crystal obtained by the Bridgman technique with seed. The crystal growth rate was 2 mm/min and the length of the single crystal was 3.5 cm. The photograph was taken at 1 cm from the end with a magnification of ×24.

The morphology of the solidification interface has been a very important subject for investigation by many scientists. The primary spacing $\lambda_1$ is one of the parameters which can correlate the growth conditions with the resulting microstructures. Most of the theoretical studies reach the analogical result, which gives:[9]

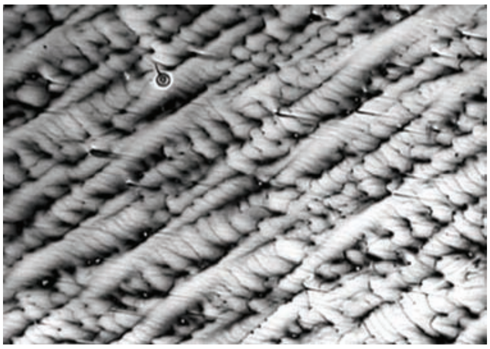
$$\lambda_1 = AG^{-m}R^{-n} \tag{1}$$

where $G$ is the temperature gradient and $R$ the crystal growth rate. $A$ is a parameter that depends on the alloy composition, while $m$ and $n$ are 0.5 and 0.25, respectively. An analysis of the primary dendrite spacing ($\lambda_1$) allows a better understanding of the mechanism of crystal growth. For the Bridgman technique, for a given growth condition, the velocity of 2 mm/min is the velocity of spontaneous nucleation. On doubling the crystal growth rate from 2 to 4 mm/min, the primary dendrite spacing remains the same value (295 μ), although the velocity has a higher value. This result is in accordance with the result from the literature.[3] On further increasing the growth rate, almost the same result was obtained, although the authors stated that some larger dendrites of the array became unstable and the tertiary arms of these dendrites grew to form new primary dendrites, thereby decreasing the average primary spacing. It is supposed that this phenomenon cannot be observed as the present system was more complicated and many motions in the system were smothered. In addition, the Bridgman method with spontaneous nucleation additionally smothers the system.

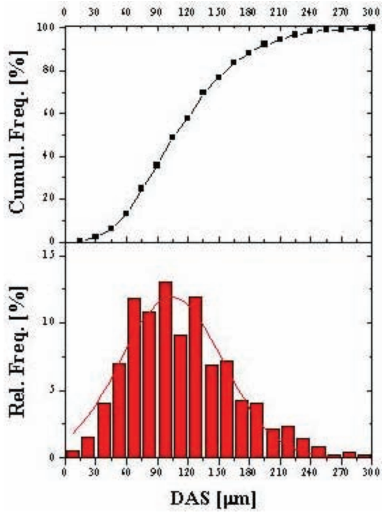
Applying the Bridgman technique with seed, the effect of an increasing change of the average primary spacing was observed when the crystal growth rate of the same crystal was changed from 11 to 2 mm/min. The differences from 125 to 288 μ can be explained if the dendrites, after an unstable period, form a stable dendrite configuration. An attempt was made to decrease the primary dendrite spacing by employing a crystal growth rate to 1 mm/min, no interesting differences could be observed and the value of $\lambda_1$ given above is the lower limit,

$\lambda_{1,min}$. The same sample grown at 6 and 2 mm/min is shown in Figs. 2 and 3, respectively. No significant differences in the primary dendrite spacing could be observed, probably due to the properties of the system (more complicated than that used in the literature).[3]

An automatic device for quantitative picture analysis, Quantimet 500 MC, and the linear measuring method were applied to the samples of the superalloy, whereby the dendrite arm spacing (*DAS*) and the distribution of *DAS* values (histograms) were obtained. The obtained results are shown in Figs. 4–8.

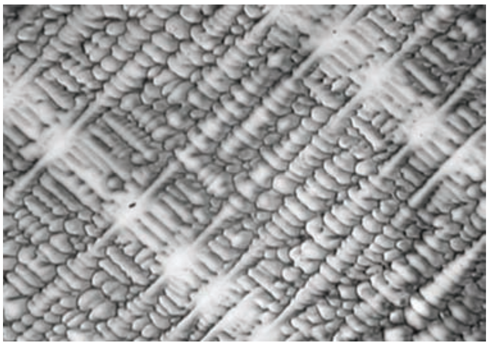


(a)




(b)

Fig. 4. a) Photograph showing the secondary dendrite spacing in a Ni-based superalloy single crystal obtained by the Bridgman technique with spontaneous nucleation and b) the histogram of the distribution of the secondary dendrite spacings. The crystal growth rate was 2 mm/min and the magnification ×20.



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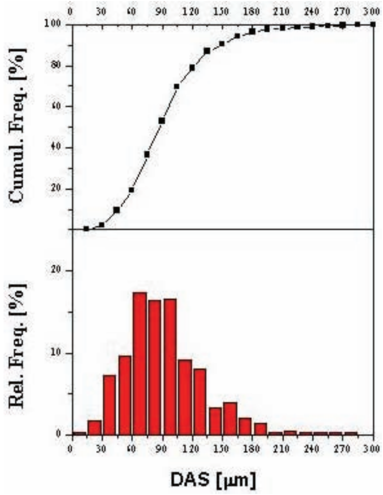
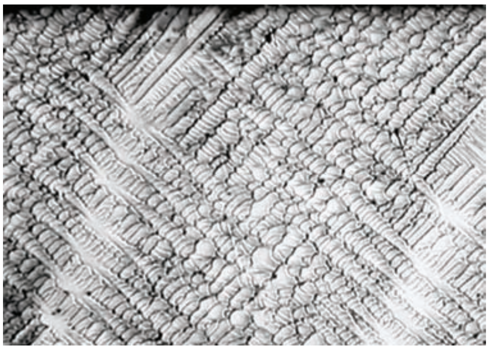
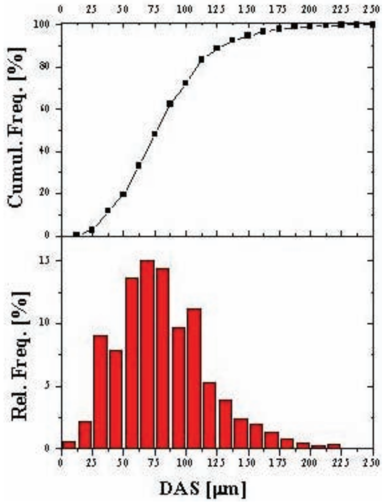



(b)

Fig. 5. a) Photograph showing the secondary dendrite spacing in a Ni-based superalloy single crystal obtained by the gradient method and b) the histogram of the distribution of the secondary dendrite spacing. The crystal growth rate was 3.4 mm/min and the magnification ×20.



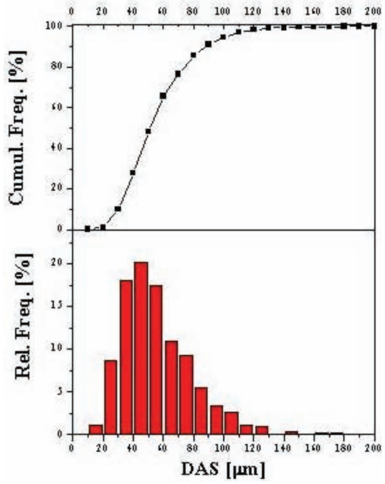
(a)




(b)

Fig. 6. a) Photograph, showing the secondary dendrite spacing in a Ni-based superalloy single crystal obtained by the Bridgman technique with spontaneous nucleation and b) the histogram of the distribution of the secondary dendrite spacing. The crystal growth rate was 4 mm/min and the magnification ×20.

(a)




(b)

Fig. 7. a) Photograph showing the secondary dendrite spacing in a Ni-based superalloy single crystal obtained by the Bridgman technique with seed and b) the histogram of the distribution of the secondary dendrite spacing. The crystal growth rate was 6 mm/min and the magnification ×20.

In accordance to Fleming's investigation,[2] the secondary dendrite spacing is a function of the temperature gradient and the crystal growth rate. This type of Relation can be express as:

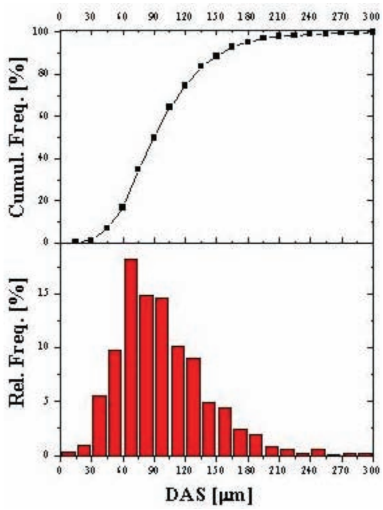
$$\lambda_2 = C(GR)^{-n} \qquad (2)$$

where $\lambda_2$ is the secondary dendrite spacing, $C$ a constant, $G$ the temperature gradient, $R$ the crystal growth rate and $n = 1/3$.

The results obtained from the Quantimet 500 MC measurements are summarized in Table I and Fig. 9 represents the linear fit for the Relation (2).

(a)

Fig. 8. a) Photograph showing the secondary dendrite spacing in Ni-based superalloy single crystal obtained by the Bridgman technique with spontaneous nucleation and b) the histogram of the distribution of the secondary dendrite spacing. The crystal growth rate was 11 mm/min and the magnification ×20.




(b)

TABLE I. Secondary dendrite spacing values of Ni-based superalloy single crystals obtained from Quantimet 500 MC measurements

| Growth method | Crystal growth rate, mm/min | Minimum distance, mm | Maximum distance, mm | Average distance, mm | Distinction % |
|---|---|---|---|---|---|
| Bridgman spont. nucleation | 2 | 0.003 | 0.314 | 0.114 | 1.351 |
| Gradient method | 3.4 | 0.006 | 0.291 | 0.084 | 1.810 |
| Bridgman spont. nucleation | 4 | 0.018 | 0.328 | 0.064 | 2.036 |
| Bridgman, with seed | 6 | 0.014 | 0.178 | 0.075 | 1.662 |
| Bridgman spont. nucleation | 11 | 0.016 | 0.238 | 0.063 | 1.996 |

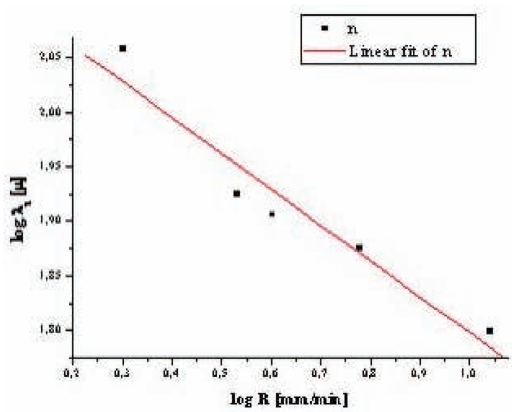



Fig. 9. Linear fit of *n* for the Relation $\lambda_2 = C(GR)^{-n}$.

It is clearly seen from Fig. 9 that n is very close to 1/3, which is in accordance to Fleming's investigation[2] that the secondary dendrite spacing is function of the temperature gradient and the crystal growth rate. In the present experiments, the temperature gradient was fixed for both Bridgman techniques but the crystal growth rate was changed.

## CONCLUSIONS

1. Single crystals of Ni-based superalloys were grown by the gradient method and by the Bridgman technique (spontaneous nucleation or from a polycrystalline seed). Comparing the applied techniques, the largest single crystal part of a sample superalloy was obtained by the Bridgman technique with spontaneous nucleation.

2. For the given growth conditions for the Bridgman technique, 2 mm/min is the velocity of spontaneous nucleation.

3. The primary dendrite spacing remained constant (295 μ), although the velocity changed from 2 mm/min to 4 mm/min, which is in accordance with published data.[5]

4. The results for the secondary dendrite spacing, obtained by Quantimet 500 MC measurements, confirmed the presumption that the secondary dendrite spacing is a function of the temperature gradient and the crystal growth rate.

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## ИЗВОД

## ПРОСТИРАЊЕ ПРИМАРНИХ И СЕКУНДАРНИХ ДЕНДРИТА МОНОКРИСТАЛА СУПЕРЛЕГУРЕ НА БАЗИ НИКЛА

СЛОБОДАНКА КОСТИЋ[1], АЛЕКСАНДАР ГОЛУБОВИЋ[1] и АНДРЕЈА ВАЛЧИЋ[2]

[1]*Институт за физику, Прегревица 118, п.пр. 68, 11080 Земун и* [2]*Технолошко–металуршки факултет, Карнегијева 4, 11000 Београд*

Монокристали суперлегуре на бази никла су расли помоћу различитих метода (градијентна метода и техника по Бриџману са спонтаном нуклеацијом и са клицом). У свим експериментима раста кристала по техници Бриџмана температурни градијент дуж вертикалне осе пећи је био константан (*G* = 33,5 °C/cm). Добијени монокристали су сечени, механички и хемијски полирани, и хемијски нагризани. Примарни и секундарни дендрити су испитивани коришћењем металографског микроскопа. Простирања крака дендрита (*DAS*) су одређивана коришћењем квантометра Quantimet 500 MC. Добијени резултати су дискутовани и упоређени са подацима из литературе.

(Примљено 5. маја, ревидирано 11. јула 2008)

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