DOI: 10.1007/s41230-016-5124-9

Effects of grain refinement on cast structure and tensile properties of superalloy K4169 at high pouring temperature

Zi-qi Jie [1], Jun Zhang [1], *Tai-wen Huang [1], Lin Liu [1], Hai-jun Su [1], Yan-li Shi [2], and Heng-zhi Fu [1]

1. State Key Laboratory of Solidification Processing, Northwestern Polytechnical University, Xi'an 710072, China

2. Xi'an Jiaotong University City College, Xi'an 710072, China

Abstract: In order to improve the filling ability of large complex thin wall castings, the pouring temperature should be increased, but this will result in the grain coarsening. To overcome this problem, two kinds of grain refiners of Co-Fe-Nb and Cr-Fe-Nb ternary alloys, which contain high stability compound particles, were prepared. The effects of the refiners on the as-cast structures and tensile properties of the K4169 superalloy with different casting conditions were studied by analyzing specimens 110 mm long and 20 mm in diameter. Results showed that the mixture addition of the two refiners in the melt of K4169 can reduce the columnar grain region and decrease the equiaxed grain size greatly. After refinement, the amount of Laves phase decreases and its morphology changes from island to blocky structure. The carbides in the fine grain samples are fine and dispersive. Meanwhile, the porosity in specimens is decreased due to grain refinement. As a result, the yield strength, ultimate strength and the elongation of the specimens are increased. The grain refinement mechanisms are also discussed.

Key words: superalloy; K4169; grain refinement; tensile properties

CLC numbers: TG143.9 **Document code:** A **Article ID:** 1672-6421(2016)02-101-06

With the continuous improvement of the engine thrust-weight ratio, the turbine disk and the intermediate case in the turbine engine become more complex in structure. Meanwhile, the operating temperature of these superalloy castings reaches approximately 700 °C. Under such a high temperature, a uniform and fine-grained microstructure is desirable in order to obtain good low cycle fatigue resistance and high tensile strength [1, 2]. Therefore, the production of such components challenges metallurgists and requires the development of advanced casting technology. One of the effective methods of improving the filling ability of large-size thin-wall castings is to increase the pouring temperature, but this will lead to a coarse grain size and decrease the low cycle fatigue resistance and tensile strength. The addition of the grain refiners in the melt is an effective way to overcome the problem and to get fine grain size [1].

Grain refinement of as-cast structure means increasing the heterogeneous nucleation sites during the solidification of castings. Fine grain casting techniques of superalloys mainly include the thermal control method [3], the chemical approach [1, 4, 5] and the dynamic method [6-8]. Among these, the chemical fine grain process is an efficient method, where the heterogeneous nucleation can be increased by the addition of a specially designed master alloy, which contains suitable solid particles with high stability in the melt. This method needs neither complicated equipment nor complex process. Refractory metal oxides, carbides, nitrides and boron have been used as refiners in some superalloys [1, 9]. However, this kind of refiner will introduce inclusions in the castings, which may become crack initiation sites, and deteriorate the mechanical properties [10]. Especially, the addition of boron will decrease incipient melting temperature of the alloys, which reduces the plastic properties greatly [11]. Liu et al [1] developed two kinds of refiners Co-Fe-Nb and Cr-Mo-Nb used in Ni-Fe based superalloys. These refiners possess effective refinement capability without introducing inclusions. However, Co-Fe-Nb and Cr-Mo-Nb can only be used at temperatures of 1,360–1,420 °C, far below the melting and pouring temperatures for most superalloy castings.

*** Tai-wen Huang**

Male, born in 1975, Ph. D., Professor. His research interests mainly focus on superalloys.

E-mail: taiwen_h@nwpu.edu.cn

Received: 2015-11-27; Accepted: 2016-01-18

Ni-based superalloy K4169 is widely used in turbine disk and intermediate case components, due to its high-temperature mechanical properties in addition to an excellent corrosion resistance [12, 13]. While the shape of these components tends to become more complex and thin-walled, leading to the bad filling. In order to improve the filling ability of complex and thin-walled castings, the pouring temperature should be increased, but this will result in grain size coarsening. To obtain good filling ability and fine microstructure, two inter-metallic compounds Co_3FeNb_2 and CrFeNb were prepared as refiners of the K4169 alloy. The constituent elements are also the elements presented in the superalloy K4169, ensuring that the grain refiners do not introduce inclusions and pose any harmful influence on the mechanical properties of the alloy. The effect of grain refiner on cast structure and tensile properties of K4169 at the pouring temperatures of 1,470–1,520 °C was investigated.

1 Materials and experimental procedure

Two kinds of ternary alloy grain refiners with the nominal compositions of Co_3FeNb_2 and CrFeNb were designed and the button ingots of the refiners were prepared by melting an appropriate proportion of the constituents in an arc melting furnace in an argon atmosphere. The raw materials were 99.95% Cr powder, 99.9% Co powder, 99.5% Fe powder and 99.97% Nb block. They were ground into powders with a size of 60–100 μm. The physical and crystallographic parameters of the refiners are listed in Table 1. The melting point of the refiners was analyzed by differential thermal analysis (DTA). The mixture of the two refiners was prepared by mixing physically with the proportion of 1:1 in weight percentage.

Table 1: Physical and crystallographic parameters of experimental refiners

Refiner	Crystal structure	Density ($g\cdot cm^{-3}$)	Melting point (°C)
Co_3FeNb_2	Hexagonal	8.8	1,550
CrFeNb	Hexagonal	8.2	>1,650

The commercial K4169 alloy with the composition (wt.%): 0.056 C, 0.01 Co, 52.54 Ni, 19.15 Cr, 3.11 Mo, 0.61 Al, 0.94 Ti, 5.03 Nb, 0.0026 B, 0.028 Zr with the balance being Fe was used for the grain refinement experiments. The equilibrium liquidus and solidus temperatures of the alloy are 1,349 and 1,270 °C, respectively, according to DTA results.

A vacuum melting furnace was used to cast ingots of K4169 superalloy. The melt was first superheated up to 1,550 °C and held for 2–4 min and then cooled down to the pouring temperature. For the conventional cast samples, the melt was poured into the preheated mold directly. However, for chemical grain refinement samples, the refiner was added into the melt at the pouring temperature. The addition amount was 0.3wt.% of the charge. After that, the melt was stirred for the refiner particles to be dispersed in the melt uniformly. Then the melt was held for 30–60 s for homogenization of the refiner and subsequently poured into the mould. The ceramic moulds with inner size of 120 mm in length and 20 mm in diameter and the preheating temperature of 900 °C were used in all cases.

The as-cast ingots were sectioned along the cross-section and the samples were ground, polished and subsequently chemically etched with a solution etchant of 15 g $CuSO_4$, 3.5 ml H_2SO_4 and 50 ml HCl to expose grain structures. The average equiaxed grain size and fraction of equiaxed grains at transverse cross-section were determined by a standard quantitative metallographic technique. The grain size was measured by the line intercept method and estimated with reference to the ASTM standard. The distribution of the alloying elements was determined using Electron-probe microanalysis (EPMA). The tensile tests were conducted using an Instron 3382 testing machine at room temperature. At least three identical specimens were tested for each case.

2 Results

The structures of transverse sections of different as-cast samples are shown in Fig. 1. For the samples without grain refinement, only columnar grains were observed. By adding the refiners, an equiaxed grain region was formed and the average grain sizes were reduced. For the samples with the pouring temperature of 1,520 °C, after the addition of mixture refiners of the two ternary

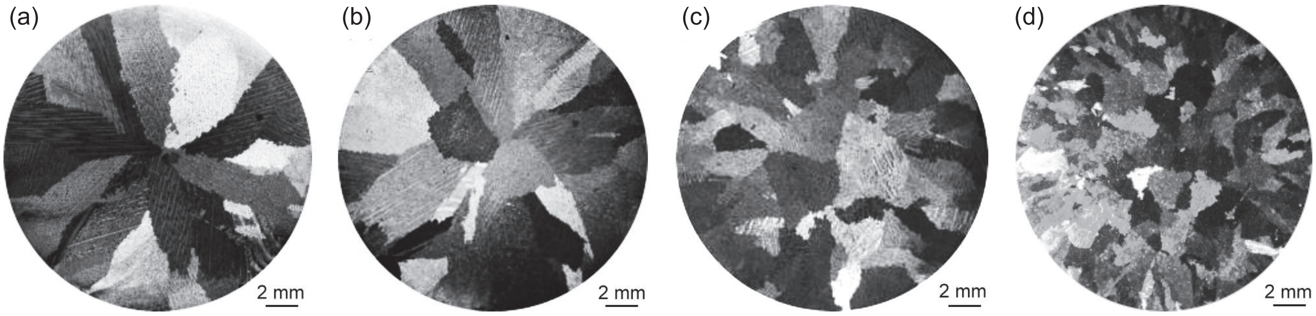

Fig. 1: Grain structures for different treatment conditions and pouring temperatures: (a) without refiner addition, 1,520 °C, (b) without refiner addition, 1,470 °C; (c) with refiner addition, 1,520 °C; (d) with refiner addition, 1,470 °C

inter-metallic compounds, the average grain size was refined from 10.56 to 2.84 mm and the proportion of equiaxed grains at cross-section was increased from 10% to 81%. Similar results were obtained for the samples with the pouring temperature of 1,470 °C, where the average grain size was refined from 8.98 to 1.85 mm and the proportion of equiaxed grains was increased from 15% to 93%.

The dendritic morphologies are shown in Fig. 2. It can be seen that the dendritic morphologies with highly developed branches were obtained in the case without grain refiner addition. However, the average length of the primary dendrite axes decreases with the addition of the grain refiners. The secondary dendrite arm spacing (SDAS) of both samples (a) and (b) was about 65 μm, and for (c) and (d) was about 58 μm, indicating that the grain refinement has a negligible effect on the SDAS, but the SDAS decreases with the decrease of the pouring temperature.

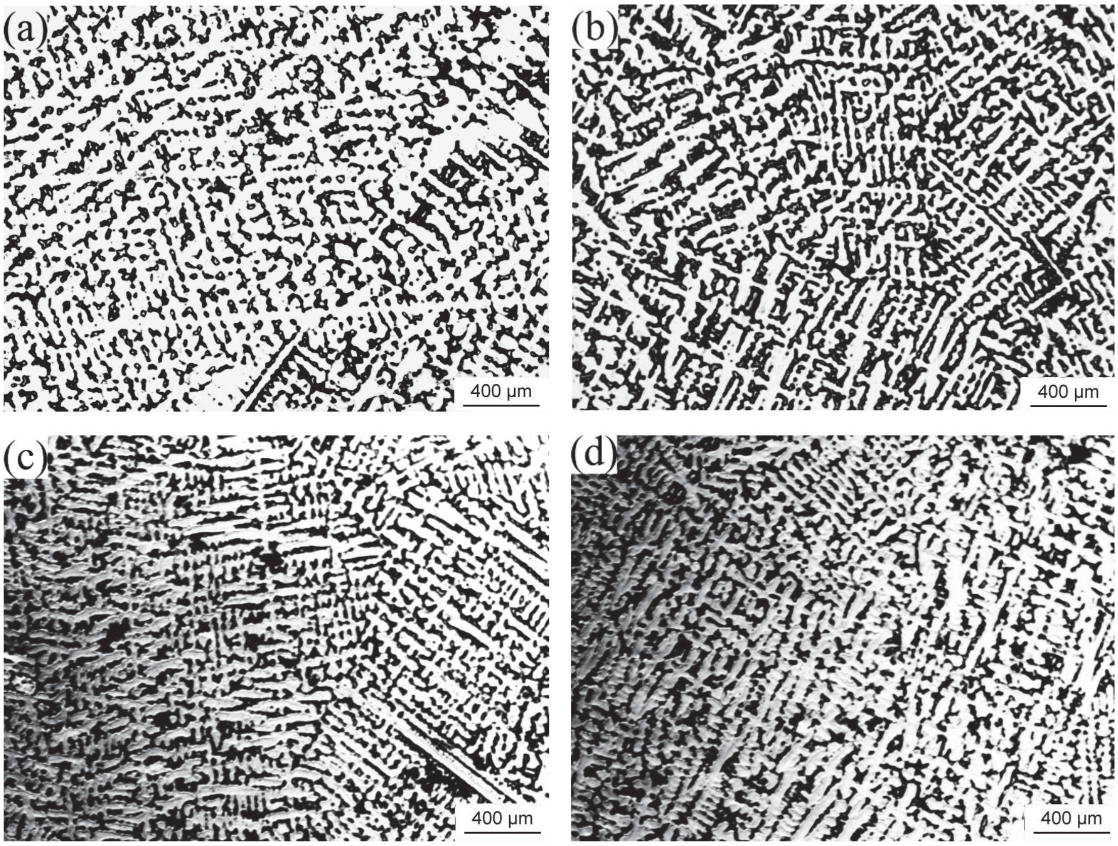

Fig.2 : Dendritic morphologies for different casting conditions: (a) without refiner addition, 1,520 °C; (b) with addition, 1,520 °C; (c) without refiner addition, 1,470 °C; (d) with addition, 1,470 °C

K4169 superalloy has a wide solidification temperature range. Therefore, porosity is likely to form in its castings. Figure 3 shows the morphology and the distribution of porosity in the samples with the grain sizes of 10.56 mm and 1.85 mm. It was shown that there exist some intensively distributed large-sized porosities in coarse grain samples. However, it becomes uniform and much smaller in the chemically refined specimen.

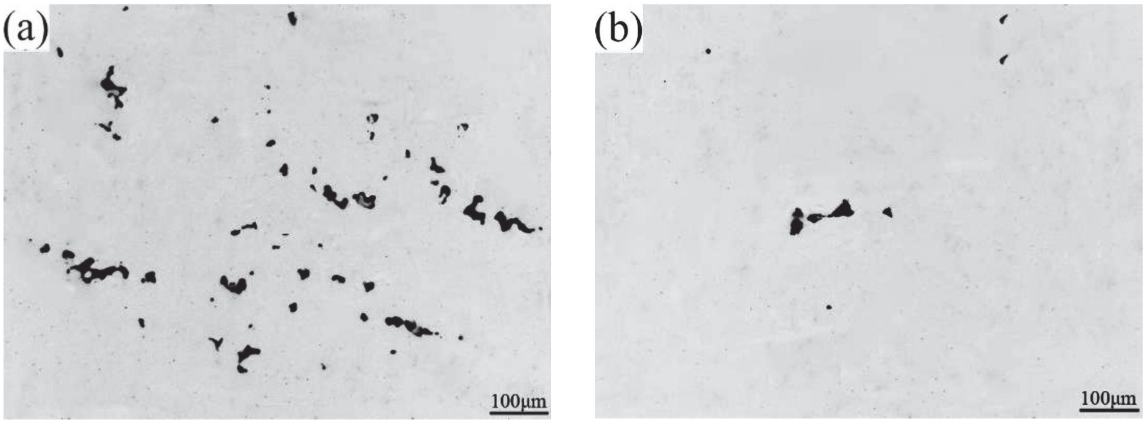

Fig. 3: Porosity of samples with different casting conditions: (a) grain size, 10.56 mm; (b) grain size, 1.85 mm

The typical as-cast microstructure of K4169 consists of the primary gamma phase dendrites, carbides, laves and delta phase [13]. Micrographs of laves and MC carbides in test bars of different grain sizes were obtained. In the test bars with the grain size of 10.56 mm and 2.84 mm, block carbides and eutectic laves can be observed. The carbide morphology in the fine grain samples is fine and dispersive. However, the laves phase is mainly contained in the eutectic phase in the coarse grain samples, and the block laves are found in the fine grain samples. The quantity of laves decreases with the decrease of grain size. Besides, the quantity of carbides remained about the same for the same pouring temperature. The results in Fig. 4 show that the volume fraction of laves phase is about 3.35% when the grain size is 10.56 mm. It was reduced to 1.48 % if the grain is refined to 2.84 mm.

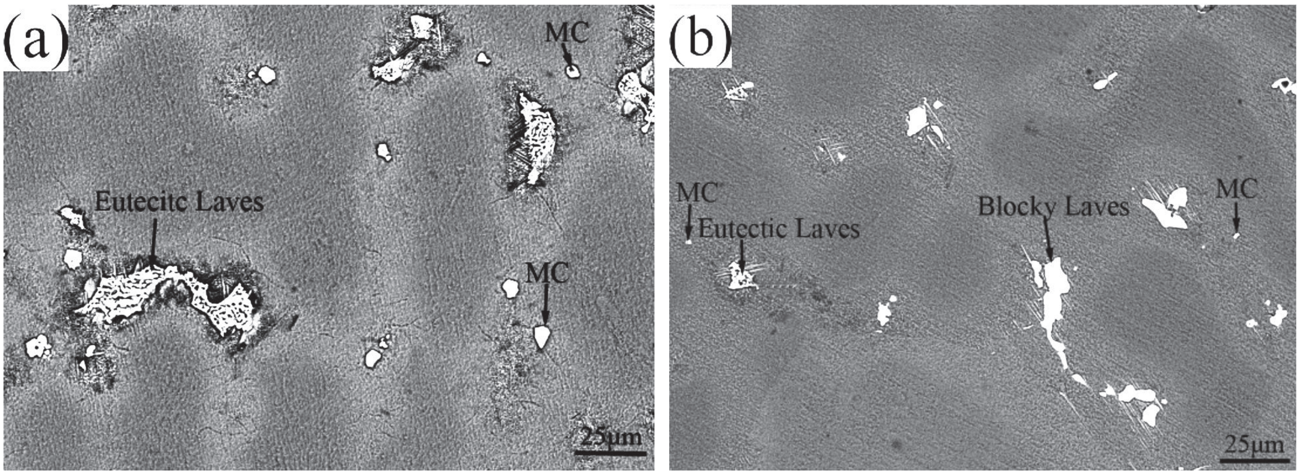

Fig. 4: Microstructure of alloy with different casting conditions: (a) without grain refiner addition, 1,520 °C, grain size 10.56 mm, (b) with refinement, 1,520 °C, grain size 2.84 mm

Figure 5 shows the correlation of grain size and ultimate tensile strength and yield strength obtained at the room temperature tensile tests. The ultimate tensile strength and yield strength of K4169 superalloy are significantly improved along with the grain refinement. When the grain size of K4169 superalloy is decreased from 10.56 mm to 2.84 mm at the pouring temperature of 1,520 °C, the tensile and yield strength are increased by 11.76% and 9.8%, respectively. For the pouring temperature of 1,470 °C, the tensile and yield strength are increased by 19.07% and 29.16%, respectively, corresponding to the grain size decreases from 8.98 mm to 1.85 mm. In addition, the elongation is increased with the addition of grain refiners at different pouring temperatures. At the pouring temperature of 1,520 °C, when the grain size is refined from 10.56 mm to 2.84 mm, the elongation is increased by 53%. When the pouring temperature is 1,470 °C, the elongation is increased by 38% corresponding to the grain size decreases from 8.98 mm to 1.85 mm.

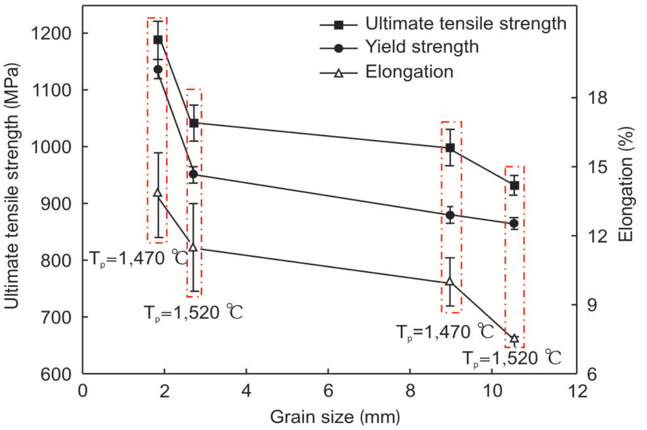

Fig. 5: Relationship between strength, elongation and grain size in K4169 superalloy

3 Discussion

Results of this study show that the refiners can lead to grain refinement and increase the proportion of equiaxed grains. The main principle is a fine epitaxial fit between low-index planes of the heterogeneous nucleation particle substrate offered by the grain refiners and the nucleated solid phase. The lower the lattice disregistry, the more effective the refiner will be in promoting nucleation. According to the calculation model of lattice disregistry (δ) between refiners and the nucleated phase proposed by Bramfitt [14], when the value of δ for some specific crystal planes is less than 12%, the refiner will have a good refining effect. Wang et al [15] calculated and simulated the planes matching models and matching orientations. The results show that (0001) and (0110) planes of refiners Co_3FeNb_2 and CrFeNb have a fine crystallographic matching relationship with the (110), (111) planes of γ matrix of K4169. Therefore, the refiners can act as the nucleation substrate of γ matrix and allow its epitaxial growth. Presence of a great number of active refiner particles in the melt would cause enormous heterogeneous nuclei of crystallites, which would impinge on one another and restrict further growth. Hence, the formation of numerous nuclei and the restriction on their further growth result in the refinement of grains. However, due to the higher pouring temperature, the refining effect is reduced.

It can be seen from Fig. 1 that adding refiner to the melt makes the equiaxed fraction increase along with the grain refinement. The addition of refiner is beneficial for forming the equiaxed grain zone. Additionally, refiner particles dispersed uniformly in the melt causes a large quantity of equiaxed grains formation. The growth of these nuclei will release a great amount of latent heat, which prohibits their further growth. In addition, the formation and growth of many equiaxed grains impede the growth of columnar grains.

The decrease of porosity in the specimen with grain refinement is due to the fact that the alloy flow distance is increased with grain refinement. The fluidity of two different conditions is tested by spiral fluidity. The fluidity is 360 mm at the condition of coarse grain and that of the fine grain is 371 mm. Dahle et al [16] also reported that finer grain size should improve fluidity of molten aluminum. This is due to grain refinement postponing dendrite coherency.

The important consequence of the solidification in superalloy K4169 is the segregation of Nb and the formation of Laves phase. Laves phase is a brittle inter-metallic topologically close-packed phase with hexagonal structure, known for its detrimental effect on mechanical properties at room temperature [17]. The main reason of laves formation is Nb and Ti segregation [17]. Figure 6 shows the correlation of grain size and segregation ratios in the K4169 superalloy. The segregation ratio is defined as the average concentration in the dendritic core over the average concentration in inter-dendritic region. The segregation ratio close to 1 indicates that the elements can reduce segregation. It can be clearly seen that the segregation of Al, Mo, Cr and Fe has little change, whereas the segregation of Nb and Ti decreases with decreasing grain size. So, this is the most important reason for the decreasing of the quantity of Laves phase.

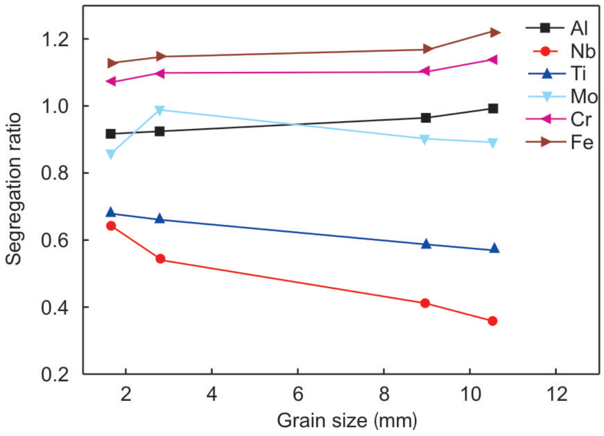

Fig. 6: Relationship between dendrite segregation ratio and grain size

The increase of the mechanical properties at room temperature in the grain refined samples is mainly due to the increase of the grain boundaries, which inhibits dislocation slide, and increases the yield strength and ultimate strength. At room temperature, the strength of the grain boundary is higher than that of the grain interior [2, 7, 18]. Therefore, the crack propagation would be impeded when encountering a grain boundary. The carbides and Laves phase in fine-grain castings are smaller than those in the coarse grain, which can also increase the yield and ultimate strength. However, high density and large size of micro-porosities in the coarse grain samples will lead to the test bars premature fracture, and cause low elongation and ultimate tensile strength.

4 Conclusions

The effects of the refiners on the as-cast structures and tensile properties of K4169 superalloy were studied. The results are summarized as follows:

(1) When adding mixed refiner of Co_3FeNb_2 and CrFeNb to the melt of K4169 superalloy, the equiaxed grain size could be refined and the proportion of equiaxed grains at cross-section could be increased in the samples with pouring temperature of 1,470–1,520 °C. Refiner particles with good lattice compatibility with matrix act as substrata of matrix, thereby causing grain refinement.

(2) As the grain refines, the amount of Laves phase decreases and its morphology changes from island to blocky structure. The carbides in the fine grain samples are fine and dispersive.

(3) The amount of porosity in the specimen could be reduced greatly after grain refinement due to the alloy flow distance being increased with grain refinement.

(4) Yield strength and ultimate tensile strength at room temperature increases significantly due to grain refinement. When the grain size of K4169 superalloy is 1.85 mm, the highest tensile and yield strength obtained are 1,189.32 MPa and 1,138.47 MPa, respectively.

References

[1] Liu Lin, Huang Taiwen, Xiong Yuhua, et al. Grain refinement of superalloy k4169 by addition of refiners: cast structure and refinement mechanisms. Materials Science and Engineering A, 2005, 394: 1–8.

[2] Du Beining, Yang Jinxia, Cui Chuanyong, et al. Effect of grain refinement on the microstructure and tensile behavior of K417G superalloy. Materials Science and Engineering A, 2015, 623: 59–67.

[3] Ma Yue, Sun Jiahua, Xie Xishang, et al. An investigation on fine-grain formation and structural character in cast IN718 superalloy. Journal of Materials Processing Technology, 2003, 137: 35–39.

[4] Xiong Yuhua, Wei Xiuying, Du Jun, et al. Grain refinement of superalloy IN718C by the addition of inoculants. Metallurgical and Materials Transaction A, 2004, 35(7): 2111–2114.

[5] Liu Lin, Zhang Rong, Wang Liuding, et al. A new method of fine grained casting for nickle-base superalloys. Journal of Materials Processing Technology, 1998, 77: 300–304.

[6] Ma Xiaoping, Li Yingju and Yang Yuansheng. Grain refinement effect of pulsed magnetic field on solidified microstructure of superalloy IN718. Journal of Materials Research, 2009, 24(10): 3174–3181.

[7] Wei C N, Bor H Y, Ma C Y, et al. A study of IN713LC superalloy grain refinement effects on microstructure and tensile properties. Materials Chemistry and Physics, 2003, 80(1): 89–93.

[8] Jin Wenzhong, Bai Fudong, Li Tingju, et al. Grain refinement of superalloy IN100 under the action of rotary magnetic fields and inoculants. Materials Letters, 2008, 62(10–11): 1585–1588.
[9] Bashir S and Thomas M C. Effect of interstitial content on High-temperature fatigue crack propagation and Low-cycle fatigue of alloy 720. Journal of Materials Engineering and Performance, 1993, 2(4): 545–550.
[10] Miao Jiashi, Pollock T M and Jones J W. Crystallographic fatigue crack initiation in Nickel-based superalloy Rene′ 88DT at elevated temperature. Acta Materialia, 2009, 57(20): 5964–5974.
[11] Liu R, Xi S Q, Kapoor S, et al. Effect of chemical composition on Solidification, microstructure and hardness of Co-Cr-W-Ni and Co-Cr-Mo-Ni alloy systems. IJRRAS, 2010, 5(2): 110–122.
[12] Li Ailan, Tang Xin, Gai Qidong, et al. Effect of heat treatment on microstructure of K4169 superalloy. Journal of Aeronautical Materials, 2006, 26(3): 311–312. (In Chinese)
[13] Li Yamin, Liu Hongjun, Liu Jie, et al. Effect of Zr addition on precipitates in K4169 superalloy. China Foundry, 2012, 9(1): 6–10.
[14] Bramfitt B L. The effect of carbide and nitride additions on the heterogeneous nucleation behavior of liquid iron. Metallurgical Transactions, 1970, 1(7): 1987–1995.
[15] Wang Fu, Zhang Jun, Huang Taiwen, et al. Preparation of inoculants used in superalloy and analysis of the atomic matching models. Journal of Materials Science & Technology, 2013, 29(4): 387–392.
[16] Dehle A K, Tondel P A, Paradies C J, et al. Effect of grain refinement on the fluidity of two commercial Al-Si foundry alloys. Metallurgical and Materials Transactions A, 1996, 27(8): 2305–2313.
[17] Janaki Ram G D, Venugopal Reddy A, Prasad Rao K, et al. Control of Laves phase in Inconel 718 GTA welds with current pulsing. Science and Technology of Welding & Joining, 2004, 9(5): 390–398.
[18] Yang Jinxia, Sun Yuan, Jin Tao, et al. Microstructure and mechanical properties of a Ni-based superalloy with refined grains. Acta Metallurgical Sinica, 2014, 50(7): 839–844. (In Chinese)

This work was financially supported by the Aeronautical Science Foundation of China (No. 2015ZE53059), the NPU Foundation for Fundamental Research (NPU-FFR-3102014JCQ01022) and the Natural fund project of the Education Department of Shaanxi Province (No. 14JK2052).