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Effect of Ru on stress rupture properties of nickel-based single crystal superalloy at high temperature

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Abstract: Two experimental single crystal superalloys, Ru-free alloy and Ru-containing alloy with [001] orientation, other alloying element contents being basically kept same, were cast in the directionally solidified furnace. The effect of Ru on the stress rupture properties of the single crystal superalloy was investigated at (980 °C, 250 MPa), (1100 °C, 140 MPa) and (1120 °C, 140 MPa). The results show that Ru can enhance high temperature stress rupture properties of single crystal superalloy. The improvement effect of Ru addition on stress rupture properties decreases with increasing test temperature. The γ' coarsening and rafting directionally are observed in Ru-free alloy and Ru-containing alloy after stress rupture test. Needle shaped TCP phases precipitated in both of alloys after stress rupture test at (1100 °C, 140 MPa) and (1120 °C, 140 MPa) and no TCP phase was observed in both of alloys after stress rupture test (980 °C, 250 MPa). The precipitate volume fraction of TCP phases is significantly decreased by the addition of Ru. At last, the relationship between the microstructure change with Ru addition and improvement of stress rupture properties was discussed.

Key words: Ni-based single crystal superalloy; Ru; stress rupture properties

1 Introduction

The demand for developing an increasingly excellent comprehensive performance of nickel-based single crystal superalloys at high temperature has resulted in an increases of the amount of refractory alloying additions [1,2]. These additions improve the high-temperature creep resistance. However, they also lead to the formation of topological close packed (TCP) phases [3,4]. The creep properties decrease as result of TCP formation within the microstructure [5,6]. It almost becomes a common sense in recent investigations that Ru additions are particularly effective in suppressing the precipitation and growth of TCP phases in single crystal superalloys [4−9]. So, the fourth-generation single crystal superalloys, such as MC-NG [10], EPM-102 [11] and TMS-138 [12], all contain certain amount of Ru element. Ru is determined to be a critical element to the success of the alloy development program as it can achieve both improved microstructural stability and increased high temperature creep strength.

Although a number of recent studies have demonstrated that the additions of Ru to high refractory content Ni-based superalloys can effectively enhance high temperature phase stability and creep resistance, the effects of Ru on the mechanisms contributing to the degradation of structural properties are not well fully understood. For instance, Ru additions also improve creep properties at relatively low temperatures, where creep properties are not determined by microstructural instabilities [13]. Therefore, in support of developing new generation single crystal superalloy, assessment of Ru addition on the comprehensive performance of the alloy must be performed. The present study examines effect of Ru on high temperature stress rupture properties of a single crystal superalloy.

2 Experimental

Pure raw materials were used in experimental study. The single crystal specimens of Ru-free alloy and Ru-containing alloy with [001] orientation were cast by means of crystal selection method in a directionally solidified furnace with high temperature gradient. The basic chemical compositions of two alloys were the same

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and all contained Cr, Co, Mo, W, Ta, Nb, Re, Al and Ni, except that the second alloy was added 2% Ru (mass fraction) substituting for Ni. The crystal orientations of the specimens were determined by Laue X-ray back reflection method, and the crystal orientation deviations of the specimens were maintained within 10° to the [001] orientation. The heat treatment of the specimens was carried out according to their corresponding heat treatment regime. The standard cylindrical specimens for stress rupture tests were machined after heat treatment. The stress rupture tests were conducted at (980 °C, 250 MPa), (1100 °C, 140 MPa) and (1120 °C, 140 MPa), respectively, in air. Microstructures of stress ruptured samples were examined. The specimens for scanning electron microscopy (SEM) observation were etched with $CuSO_4$+HCl+H_2O. Foils for transmission electron microscopy (TEM) analysis were obtained by cutting 0.2 mm-thick discs perpendicular to the tensile axis of the specimens using an electric discharge machine. Thin foils were prepared by twin-jet thinning electrolytically in a solution of 10% perchloric acid and 90% ethanol at −10 °C using liquid nitrogen. The microstructures of the alloys were examined using SEM and TEM.

3 Results

3.1 Stress rupture properties

The stress rupture properties of Ru-free alloy and Ru-containing alloy are shown in Fig. 1. It can be seen that the stress rupture life and elongation are increased with Ru addition. This indicates that addition of Ru to high refractory content Ni-based single crystal superalloy can improve high temperature stress rupture property. The stress rupture lives are increased by 24%, 17% and 14%, respectively, at different test conditions of (980 °C, 250 MPa), (1100 °C, 140 MPa) and (1120 °C, 140 MPa). Therefore, the improvement effect of Ru addition on stress rupture properties decreases with increasing test temperature.

3.2 Microstructures of stress ruptured specimens

The microstructures of the longitudinal section in the ruptured specimens of Ru-free alloy and Ru-containing alloy at different conditions were observed by SEM. Figures 2 and 3 illustrate the microstructure apart from fracture surface 2 cm and near fracture surface of the ruptured specimens, respectively.

The microstructures obtained after stress rupture tests reveal evidence of coarsening of precipitates in a direction transverse to the applied stress. The formation of such rafts is known to occur by growth of precipitates perpendicular to the direction of stress followed by their coalescence. Comparison of the microstructures shown in Fig. 2 clearly indicates that the average raft thickness

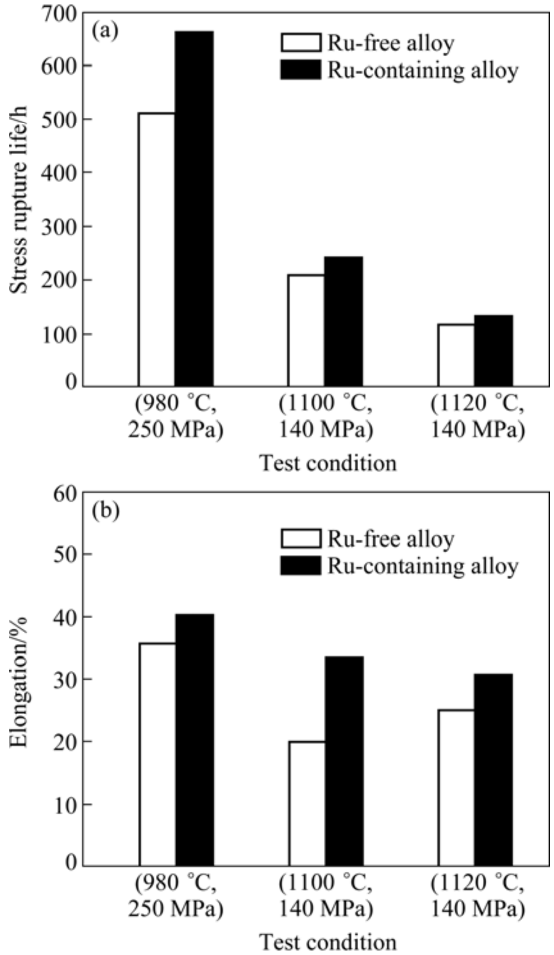

Fig. 1 Stress rupture properties of Ru-free alloy and Ru-containing alloy: (a) Stress rupture life; (b) Elongation

varied with the test conditions. The adjacent γ' particles met and fused together to produce rafts at condition of (980 °C, 250 MPa). The γ phase was no longer continuous at condition of (1100 °C, 140 MPa). Islands were forming and entirely surrounded by the γ' phase at condition of (1120 °C, 140 MPa). It is certainly intuitive that temperature of test would have a strong influence on the amount of coarsening. The thickness of γ' phase observed in microstructures increases with increasing the test temperature. Atomic diffusion controls the whole rafting process. The higher the temperature is and the faster the atoms diffuse, the faster the γ' phase grows and rafts. So, with the rise of temperature, the thickness of γ' rafts turns bigger. Moreover, it can be seen that the coarsening extent of γ' phase of Ru-free alloy is severer than that of Ru-containing alloy. The γ' rafts of Ru-containing alloy are more regular and perfect than those of Ru-free alloy.

Figure 3 illustrates the microstructure near fracture surface of the ruptured specimens. No TCP phase is observed in the ruptured specimens tested at (980 °C, 250 MPa). The specimens tested at (1100 °C, 140 MPa)

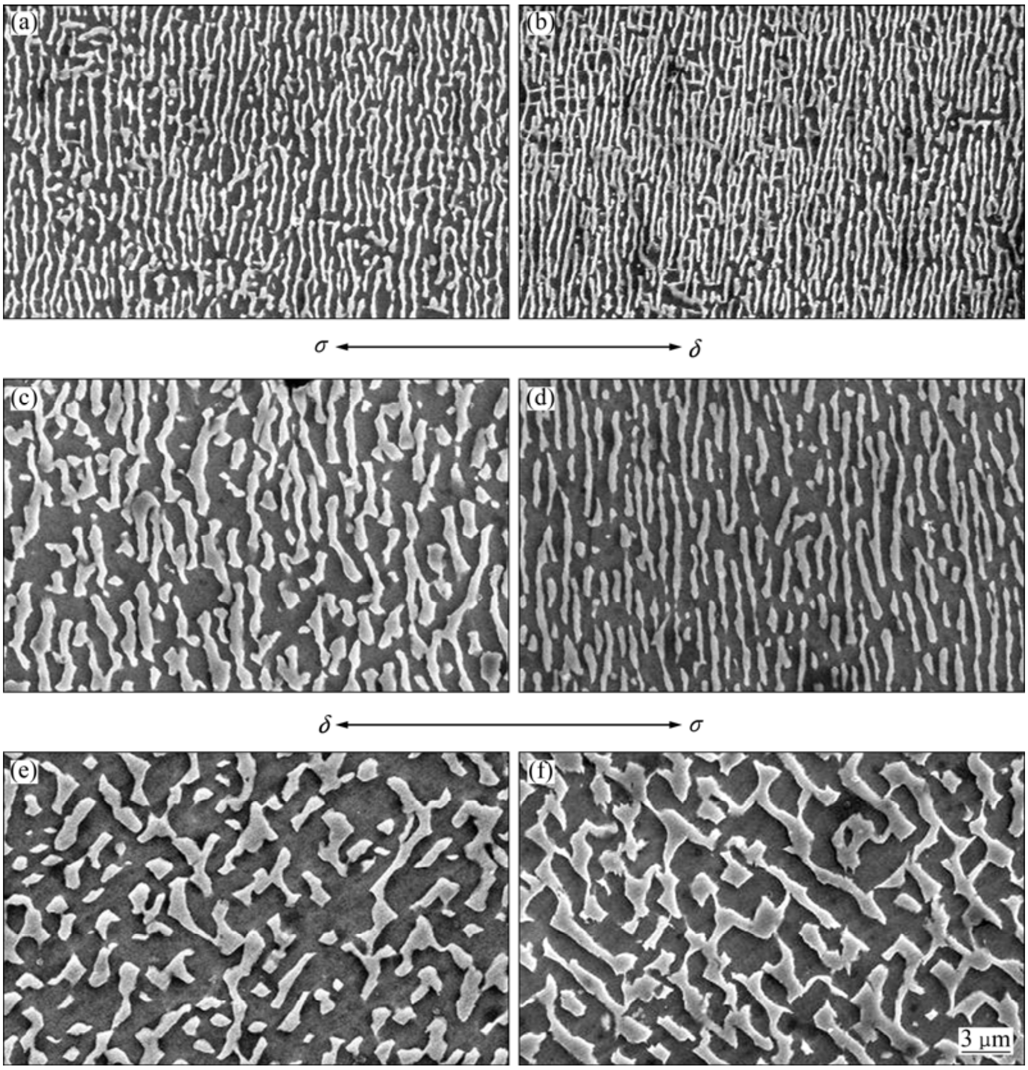

Fig. 2 Microstructures apart from fracture surface 2 cm of ruptured specimens: (a) Ru-free alloy tested at (980 °C, 250 MPa); (b) Ru-containing alloy tested at (980 °C, 250 MPa); (c) Ru-free alloy tested at (1100 °C, 140 MPa); (d) Ru-containing alloy tested at (1100 °C, 140 MPa); (e) Ru-free alloy tested at (1120 °C, 140 MPa); (f) Ru-containing alloy tested at (1120 °C, 140 MPa)

and (1120 °C, 140 MPa) exhibit the presence of TCP phases in Ru-free alloy and Ru-containing alloy. The number of TCP phases increases with increasing temperature. The cracks have formed at different test conditions in both of alloys. TCP phase is brittle and becomes the site of crack initiation, as shown in Figs. 3(c)−(f). Moreover, it has also been shown that the precipitate volume fraction of TCP phases is decreased by the addition of Ru. This indicates that Ru can enhance high temperature microstructure and phase stability of Ni-based single crystal superalloy. This is in good agreement with the result carried out in other studies [4−9].

The formation of TCP phases in Ni-based single crystal superalloys is generally attributed to the supersaturation of high melting-point refractory elements (Re, W, Mo) within the disordered γ phase [8]. As the levels of these refractory alloying elements added are increased to enhance creep properties of single crystal superalloys, the limited degree of refractory element solubility within the γ' phase increases. The addition of Ru decreases the partition ratio of TCP forming elements, Re, W, Mo [5]. So, the saturation degrees of Re, W, Mo element were decreased by Ru addition, which can enable Ru-containing alloy to have more resistance to the formation of TCP phases [4,5,8].

The chemical compositions of TCP phases in the stress ruptured specimens at (1120 °C at 140 MPa) for Ru-free alloy and Ru-containing alloy using SEM/EDS X-ray analysis are shown in Fig. 4. In contrast, the TCP phases in Ru-containing alloy contain more of Ru element compared with Ru-free alloy. Other refractory elements have similar levels in the TCP phases. In both of alloys, Re, W, Co, Mo, Cr are enriched in the TCP phases.

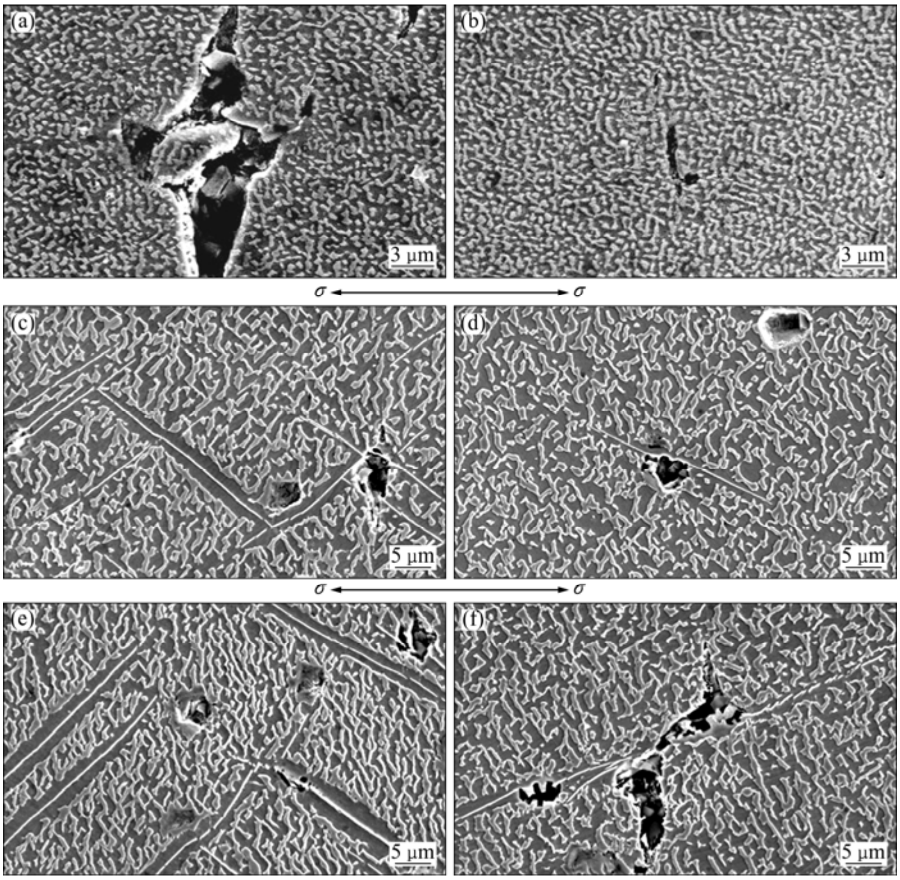

Fig. 3 Microstructures near fracture surface of ruptured specimens: (a) Ru-free alloy tested at (980 °C, 250 MPa); (b) Ru-containing alloy tested at (980 °C, 250 MPa); (c) Ru-free alloy tested at (1100 °C, 140 MPa); (d) Ru-containing alloy tested at (1100 °C, 140 MPa); (e) Ru-free alloy tested at (1120 °C, 140 MPa); (f) Ru-containing alloy tested at (1120 °C, 140 MPa)

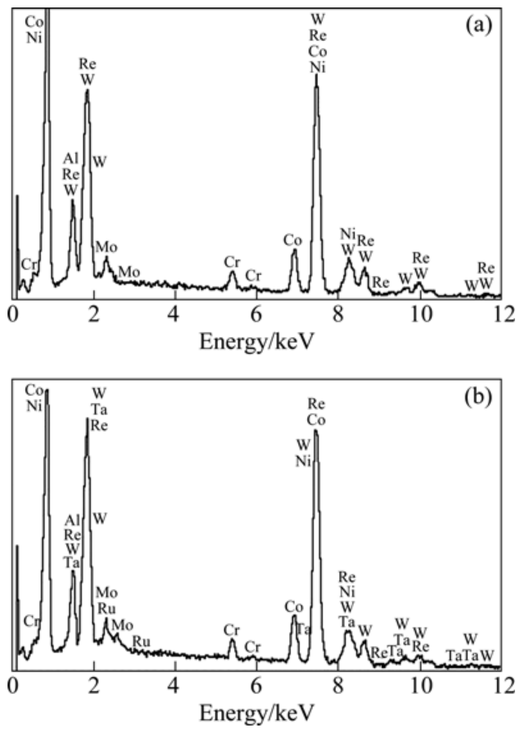

Fig. 4 EDX analyses of TCP phases in specimens ruptured at (1120 °C, 140 MPa): (a) Ru-free alloy; (b) Ru-containing alloy

4 Discussion

Ru plays an important role on microstructure and phase stability for the new generation single crystal superalloy [6−12]. The difference between Ru-free alloy and Ru-containing alloy can largely be attributed to the change of microstructure by Ru addition. At elevated temperatures, the creep resistance of Ni-based single crystal superalloys is dependent upon a number of factors including lattice misfit and microstructure stability. The results showed that each of factor is affected by the addition of Ru. The implications of these associated changes are discussed with respect to the results presented in this study.

The lattice misfit has an important relation with creep property for superalloy during high temperature creep deformation. It is well known that the lattice misfit of nickel-based superalloys is strongly influenced by the alloy composition. The alloying element Ru increases the amount of lattice misfit as it partitions preferentially to the γ matrix [6]. During creep deformation at elevated temperatures, the microstructure of Ni-based single crystal superalloys can evolve from discrete cuboidal precipitates embedded in γ matrix to coarse rafted structures, as shown in Fig. 2. Coherency stress due to the lattice misfit between *γ* and *γ*′ phases acts as the driving force for the microstructural change. The rafted structure can be an effective barrier confining dislocation activity within the discrete *γ* rafts, leading to steady state creep. It can be seen from Fig. 2 that the *γ*′ rafts in Ru-containing alloy are more regular and perfect than those in Ru-free alloy.

Moreover, the higher misfit in the Ru-containing alloy correlates well with the finer dislocation spacing and lower minimum creep strain rate [14]. Figure 5 shows the morphologies of *γ*/*γ*′ interfacial dislocation of stress ruptured specimens at (980 °C, 250 MPa). It is clear that the dislocation network in Ru-containing alloy is denser and much more than that in Ru-free alloy. The dense *γ*/*γ*′ interfacial dislocation network will effectively prevent the dislocation from cutting into *γ*′ phase in the creep process [15,16]. Therefore, this is the main reason that the stress rupture properties of Ru-containing alloy are better than those of Ru-free alloy at (980 °C, 250 MPa), where TCP phases do not precipitate and creep properties are not determined by microstructural instabilities.

It can be seen from Fig. 3 that TCP phases all precipitate in both of alloys during stress rupture test at (1100 °C, 140 MPa) and (1120 °C, 140 MPa). However,

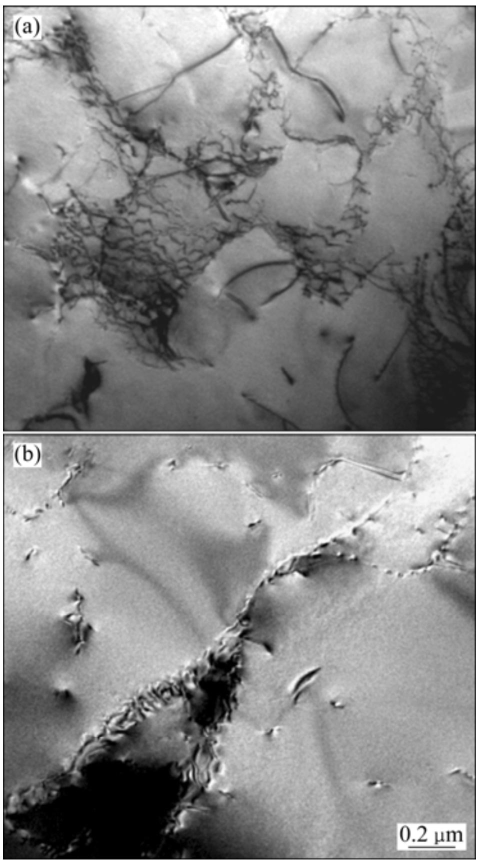

Fig. 5 TEM images of ruptured specimens tested at (980 °C, 250 MPa): (a) Ru-free alloy; (b) Ru-containing alloy

Ru-containing alloy is more resistant to the formation of TCP phases. It is another reason that Ru-containing alloy has higher stress rupture properties than Ru-free alloy at these conditions.

The microstructure stability at elevated temperatures is a key concern for the single crystal superalloys. TCP phases are detrimental to stress rupture properties of alloys [3, 4]. There are three main reasons. Firstly, TCP phases are brittle and will become the sites of crack initiation and easy ways for the crack propagation during stress rupture test. Secondly, the precipitates of TCP phases destroy the continuity of the rafted microstructure. Formation of regular and perfect *γ*′ rafts has a good effect on the stress rupture life of single crystal superalloy. It is shown in Fig. 3 that the microcracks come into being in disruption of rafted structure of *γ*′ precipitates, and high stress concentration at TCP-matrix interfaces or at points interlinking the TCP precipitates, leading to microcracking. Finally, in both of alloys, solid solution strengthening elements, such as Re, W, Co, Mo are enriched in the TCP phase, as shown in Fig. 4. When the precipitation of TCP phase occurs within the system, these solid solution strengthening elements are depleted from the surrounding matrix. Therefore, these lead to an extensive envelope of *γ*′ around the TCP which may potentially act as a channel for preferential deformation [5]. Therefore, Ru-containing alloy is more resistant to the formation of TCP phases, which can enable that Ru-containing alloy has higher stress rupture properties than Ru-free alloy at conditions of (1100 °C, 140 MPa) and (1120 °C, 140 MPa).

5 Conclusions

1) Ru can enhance high temperature stress rupture properties of single crystal superalloy. The improvement effect of Ru addition on stress rupture properties decreases with increasing test temperature.

2) Needle-shaped TCP phases precipitate in both of alloys after stress rupture test at (1100 °C, 140 MPa) and (1120 °C, 140 MPa) and no TCP phases is observed in both of alloys after stress rupture test (980 °C, 250 MPa). The precipitate volume fraction of TCP phases is significantly decreased by the addition of Ru.

3) Addition of Ru improves microstructural stability at high temperatures by decreasing the formation of TCP phases. This phenomenon indirectly improves the creep resistance because creep properties are determined by microstructural instabilities.

4) A higher misfit in the Ru-containing alloy makes more perfect rafts and denser dislocation networks, which lowers creep strain rates and improves stress rupture properties of the alloy at high temperature.

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Ru 对镍基单晶高温合金高温持久性能的影响

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摘　要：在真空定向炉中浇注了具有[001]方向的不含 Ru 和含 Ru 两个单晶高温合金，其它合金元素的含量基本相同，研究 Ru 对单晶高温合金在(980 °C，250 MPa)，(1100 °C，140 MPa)和(1120 °C，140 MPa)条件下持久性能的影响。结果表明，加入 Ru 能提高单晶高温合金的高温持久性能，提高作用随着温度的升高而降低。在断裂后的两种合金试样中都观察到 γ'相定向粗化和筏排化，并且在(1100 °C，140 MPa)和(1120 °C，140 MPa)条件试样中有针状的 TCP 相析出，而在(980 °C，250 MPa)条件试样中无 TCP 相析出。加入 Ru 减少了 TCP 相的析出数量。最后，讨论了加入 Ru 带来的合金组织变化进而提高合金持久性能的原因。

关键词：镍基单晶高温合金；Ru；持久性能

(Edited by YANG Hua)