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Refinement Mechanism of Microstructure of Undercooled Nickel Based Alloys

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Abstract: Through the use of purification and recirculation superheating techniques on molten glass, the Ni65Cu33Co2 alloy was successfully undercooled to a maximum temperature of 292 K. High-speed photography was employed to capture the process of interface migration of the alloy liquid, allowing for an analysis of the relationship between the morphological characteristics of the alloy liquid solidification front and the degree of undercooling. Additionally, the microstructure of the alloy was examined using metallographic microscopy, leading to a systematic study of the microscopic morphological characteristics and evolution laws of the refined structure during rapid solidification. The research reveals that the grain refining mechanism of the Ni-Cu-Co ternary alloy is consistent with that of the binary alloy (Ni-Cu). Specifically, under low undercooling conditions, intense dendritic remelting was found to cause grain refinement, while under high undercooling conditions, recrystallization driven by accumulated stress and plastic strain resulting from the interaction between the liquid flow and the primary dendrites caused by rapid solidification was identified as the main factor contributing to grain refinement. Furthermore, the study highlights the significant role of the Co element in influencing the solidification rate and reheat effect of the alloy. The addition of Co was also found to facilitate the formation of non-segregated solidification structure, indicating its importance in the overall solidification process.

Key words: grain refinement; recalescence; recrystallization; undercooling

1 Introduction

Rapid solidification technology began in the late 1950s and is a rapidly developing research direction in the field of materials science and engineering[1-4]. According to the different undercooling methods, rapid solidification technology can be divided into quenching rapid solidification and deep undercooling rapid solidification. The phenomenon of undercooling refers to the phenomenon that liquid metal is cooled to a temperature below the equilibrium liquid phase line without crystallization[5-8]. The undercooling tendency of the melt strongly depends on the size of heterogeneous nucleation kinetics, and there are usually two methods to obtain undercooled melts: (1) reduce or eliminate heterogeneous nucleation sites; (2) through methods such as melt overcooling, dulling the surface of heterogeneous nucleation sites to lose their heterogeneous nucleation effect. The characteristic of deep undercooled rapid solidification is that the rapid solidification is not constrained by external heat dissipation, and under a sufficiently high degree of undercooling, the entire melt can achieve rapid solidification under slow cooling conditions. In most cases, the solidification of most alloy melts is carried out through heterogeneous nucleation[9-15]. In order to achieve deep undercooling/undercooling of liquid metal, it is necessary to microscopically purify it and maximize the elimination or dulling of heterogeneous nuclei in the melt. The methods commonly used to achieve thermodynamic deep undercooling include the method of tiny droplets, the method of cyclic overheating, the method of molten glass purification, the method of electromagnetic levitation melting, and the method of falling tube.

Once the undercooled melt forms a crystal nucleus, it will solidify rapidly, and at this point, the speed at which the crystallization latent heat is released is much faster than the speed at which the system dissipates heat to the surroundings[16]. The system is reheated to a higher temperature, which is known as the recalescence phenomenon. The recalescence phenomenon ends

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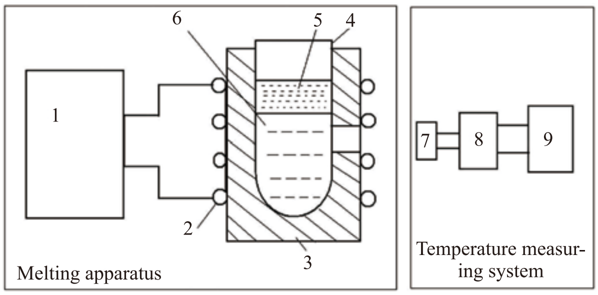
when the rapid solidification process ends, and then enters the slow cooling and solidification stage, with the residual liquid phase solidifying through heat dissipation to the surroundings. Therefore, compared with the conventional solidification process, the dendrites formed during the rapid solidification of deeply undercooled melts undergo intense physical and chemical effects, such as thermal shock and liquid flow impact. The integrity of the primary dendrites is often damaged and may even completely disappear in the final structure[17]. Experimental evidence has shown that under deeply undercooled conditions, when the degree of undercooling of the alloy melt exceeds a certain critical value, grain refinement phenomena occur in many alloy systems. Since the 1950s, research on the grain refinement mechanism in deeply undercooled alloys has been ongoing, but to date, there is still no unified mechanism that can explain the refining phenomena in all alloys.

In nonequilibrium solidification, grain refinement is widely present in various binary alloys as a representation of the microstructure of meta-stable materials, such as Fe-Co alloy, Cu-Co alloy and Ni-Cu alloy. When previous scholars studied the relationship between undercooling and structure evolution in different metallic systems, their main focus was on similar grain refinement phenomena[19-23]. Compared with previous mature theories, whether the recrystallization theory is the intrinsic mechanism in the process of undercooling solidification still needs further discussion. Throughout the experiments, we focused mainly on Ni-Cu single-phase alloy. Because Ni and Cu can form continuous solid solutions, it has a wide single-phase region and significantly better properties than other alloys, thus widely used in the industrial field[23]. At present, there is a wide range of deep undercooling experiments being conducted on Ni-Cu binary alloys, and significant advancements have been made in establishing non-equilibrium solidification models. Furthermore, the mechanism for grain refinement under rapid solidification has been thoroughly elucidated[21]. Despite these developments, the grain refinement mechanism in ternary single-phase alloys remains unconfirmed, and the impact of introducing a third element on the non-equilibrium solidification effect has not been fully investigated. Nonetheless, it is noteworthy that even a small quantity of Co has the ability to create a single-phase solid solution in Ni-Cu alloy, thus providing substantial support for our research.

This study aimed to achieve different levels of undercooling in the Ni65Cu33Co2 alloy through experimental techniques involving the purification of molten glass and loop superheat. The dynamic evolution of the solidification front was observed using a high speed camera. The resulting fine-grained structure at varying degrees of undercooling was analyzed using EBSD and TEM techniques. The objective of the research was to investigate whether the grain refinement mechanism is the same for the Ni-Cu binary single-phase alloy and the Ni-Cu-Co ternary single-phase alloy under undercooling conditions. Additionally, the study aimed to examine the effect of Co addition on the microstructure transformation of the undercooled Ni-Cu binary alloy. The findings of this study provide strong support for the theory of rapid solidification of the single phase alloy.

2 Experimental



1-high fiequency power 2-induction coils 3-substrate 4-crucible 5-melted glass 6-mlet 7-infrared probe 8-filter 9-3 056 recorder

Fig.1 Schematic diagram of in-situ melting/undercooling and thermometric apparatus

In the protective environment full of argon (Ar), pure Ni (99.99%), pure Cu (99.99%), and pure Co (99.99%) are used to melt in a vacuum arc melting furnace (Fig.1) to prepare Ni-Cu and Ni-Cu-Co master alloys with uniform composition.Use an ultrasonic cleaner containing a small amount of alcohol to clean the alloy sample and quartz tube for 30 minutes in advance to preliminarily remove impurities. Using pure B_2O_3 and NaO-SiO_2-CaO glass as purifying agents, pure B_2O_3 is placed in a ceramic crucible before use, dehydrated at low temperature, heated to 1 073 K for 5 hours, and then cooled with the furnace. The fired glass is broken and placed in a desiccator for later use. In order to ensure the purity of the sample, high-purity Fe, Co, and Cu are mechanically ground and rusted before the experiment, and then cleaned with dilute hydrochloric acid and anhydrous alcohol. The melting and purification of the experimental alloy are carried out in the same quartz crucible. To ensure that the cooling conditions of the samples are roughly the same, the weight of the sample is set to 5 g. During the experiment, the quartz crucible is first placed with pure metal blocks weighed according to the composition ratio. In order to better purify, a small amount of B_2O_3 glass is placed in the middle and bottom of the raw material, and a certain amount of fired B_2O_3 glass is covered on top. Then it is put into the induction furnace for heating and the circulation overheating and glass purification process. The softening point of B_2O_3 glass is about 600 ℃. The metal induced heating with the B_2O_3 glass in contact with it will melt first, wrapping the metal in it to avoid oxidation during melting. In order to obtain a large degree of undercooling, the purification of the alloy melt and deep undercooling rapid solidification are carried out together in the high-frequency induction furnace, and its micro-purification technology includes the following three links, namely, induction smelting + glass purification + circulation overheating purification. By implementing the method of heating-melting overheating-cooling cycle to purify the alloy liquid coated with glass, the adjustable process parameters are: overheating temperature, melting rate, holding time, and number of cycles. The specific operation is to hold at an overheat of about 100-150 K for 3-5 minutes, then add a small amount of (about 1/6 of B_2O_3 glass) NaO-SiO_2-CaO glass, and then implement overheating-cooling cycles several times to achieve deep undercooling of the alloy. This paragraph is written in Chinese. After cutting, grinding, and polishing different undercooled solidification samples, we used a mixed acid solution of 30 mL HNO_3 and 30 mL HCL for corrosion, and the corrosion time depends on the specific situation. Next, we used an optical microscope (OM, Leica DM2500M) to observe the evolution of the solidification structure and captured metallographic images. We selected samples with typical grain refinement structures for vibratory polishing to remove surface stress layers, and characterized them using electron backscatter diffraction (EBSD). We also prepared samples with the maximum undercooling using focused ion beam (FIB) milling and characterized them using transmission electron microscopy (TEM). To conduct microhardness tests on each sample within the undercooling range, we used a microhardness tester (HMV-2T) and applied a loading force of HV0.5. To ensure the accuracy of the hardness tests, we selected 20 points on the surface of each undercooled sample for testing and took the average of these test values as the final result.

3 Results and discussion

3.1 Analysis of solidification structure evolution

The phenomenon of undercooling during non-equilibrium solidification refers to the difference between the liquid phase line temperature TL and the nucleation temperature TN in the alloy equilibrium phase diagram. The degree of undercooling can be controlled using rapid solidification techniques. The rapid solidification process of alloy melts[18] can be roughly divided into four stages: pre-solidification cooling stage, recrystallization[19], heating stage during recrystallization (solidification latent heat takes effect), and post-solidification cooling stage. Fig.2 illustrates the temperature curve during the rapid solidification process. Recrystallization occurs when the alloy melt cools to the nucleation temperature T_N. Due to the release of solidification latent heat, the temperature returns to the maximum recrystallization temperature T_R, and eventually cools to room temperature. ΔT represents the degree of undercooling during the solidification process, while ΔT_R represents recrystallization and is used to indicate the degree of non-equilibrium solidification. During the recrystallization process, the proportion of primary solid phase is directly proportional to ΔT_R, *ie*, the higher the ΔT_R, the more primary solid phase will be produced during the recrystallization process, thus storing energy for the transformation of the solidified structure.

Fig.2 shows the rapid solidification curves of Ni65Cu33Co2 alloy under different undercooling conditions. It can be seen that the initial stages of undercooling and recrystallization of the binary alloy are not obvious, they are similar. With the increase of undercooling degree and the advancement of the solidification process, recrystallization increases linearly, and the slow solidification stage after recrystallization gradually shortens. The research results show that undercooling can enhance the recrystallization effect in the solidification process, and shorten the cooling and solidification time after recrystallization. By analyz-

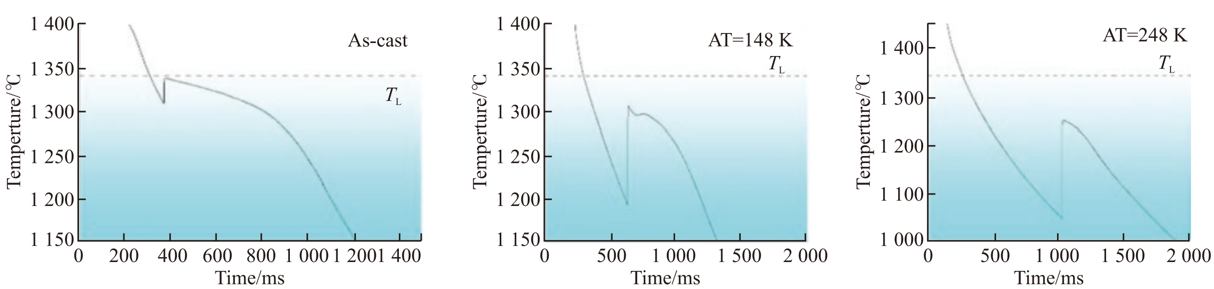


Fig.2 Rapid solidification curves of Ni65Cu33Co2 alloy with different undercoolings

ing the microstructure morphology of all undercooled samples, the microstructural evolution process of the three alloys with increasing undercooling can be seen as shown in the figure. In the initial Ni65Cu35 alloy, two types of grain refinement and different forms of solidification structure can be observed. Therefore, these results are of great significance for understanding the solidification process and microstructure evolution of alloys under undercooling conditions. At the same time, this provides guidance for the preparation and performance regulation of alloy materials, and provides important references for research and application in related fields.

As the undercooling increasing, the microstructure of Ni65Cu35 alloy experienced two grain refinement events. In the first refinement, the circular equiaxed grains gradually transformed into equiaxed grains with straight grain boundaries, while in the second refinement, the grain size became smaller. For the Ni65Cu33Co2 alloy, the evolution of the microstructure can be divided into four ranges, see Fig.3. Firstly, under very small undercooling conditions ($\Delta T < 44$ K), the microstructure of the alloy exhibited a dendritic morphology, with many secondary dendrites surrounding the primary dendrite. The length of the primary dendrite was relatively large, and the overall dendrites did not have a fixed growth direction, presenting a relatively loose structure. Secondly, in the undercooling range of 44 K < ΔT < 109 K, the dendrites began to transform into equiaxed grains, and the first grain refinement started from here. As the undercooling increasing, all the dendrite transformed into the equiaxed grain. Finally, at ΔT=109 K, equiaxed grains occupied the entire structure morphology, and oriented dendrites appeared, marking the end of the first refinement. Then, in the undercooling range of 109 K < ΔT < 177 K, the microstructure of the alloy was mainly composed of oriented fine dendrites. With the continuous improvement of undercooling, the density and orientation of fine dendrites also continued to improve. Finally, when ΔT > 177 K, all fine dendrites transformed in the direction of deformed grains, and many deformed grains appeared, marking the second grain refinement begin. As the undercooling increasing, the microstructure morphology consisted only of finer equiaxed grains and twinned grains.

3.2 The liquid-solid front during solidification

Due to the high solidification rate, the solidification process of alloy melt under deep undercooling treatment is usually not observed with specific solidification dynamics. However, by recording the re-emission phenomenon during the solidification process, dynamic images of the solidification interface can be obtained using a high-speed camera. By measuring the distance and time of the solid-liquid interface migration, the interface migration speed of the alloy at each undercooling can be calculated, that is, the solidification speed. Fig.4 shows the pre-curing image of Ni65Cu33Co2 alloy, indicating the dynamic images obtained from the re-irradiation phenomenon recorded during the curing process using a high-speed camera. These data will help enhance our understanding of the alloy curing process and provide critical references for further research and applications.

The analysis of solidification interface images of two alloys reveals distinct characteristics. Darker regions represent the uncooled alloy liquid phase (denoted as "L" in Fig.4), while the brighter areas represent the solid phase initially generated during the solidification process, which releases heat (denoted as "S" in Fig.4). At different degrees of undercooling, the dynamics of the alloy solidification interface vary significantly: at small undercooling, the solidification interface of the alloy liquid tends to form shapes with smaller angles

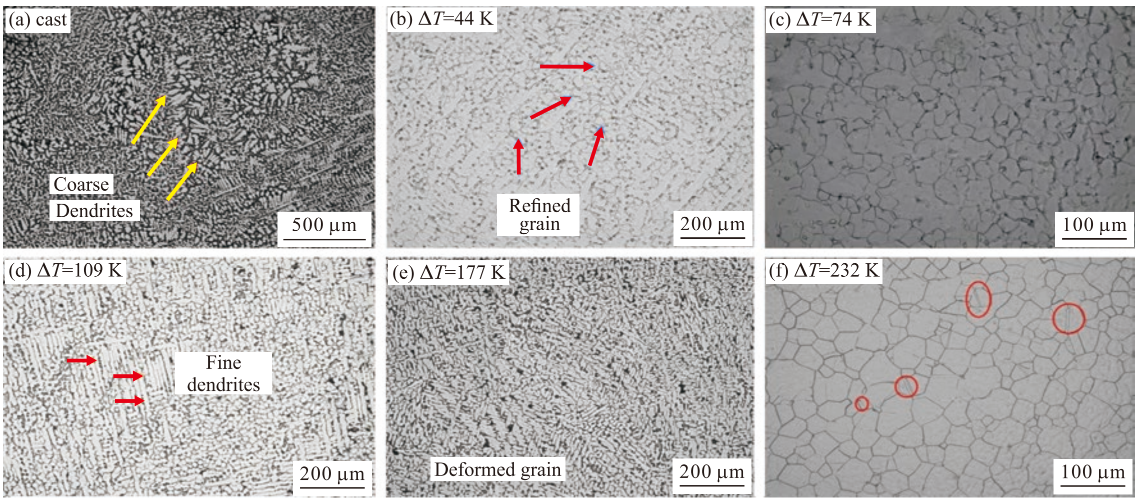


Fig.3 Evolution law of micro-structure of the Ni65Cu33Co2 alloys with undercoolings

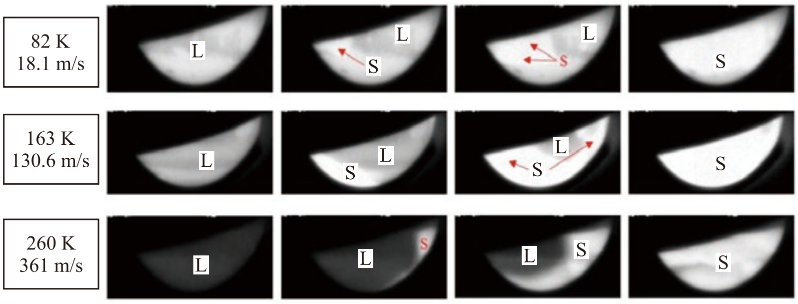


Fig.4 Solidification front evolution of the Ni65Cu33Co2 alloys with different undercoolings

in a fixed direction, where the solidification rate at each position does not differ significantly, indicating a slow stage; as the undercooling increases to a moderate level, the alloy liquid progresses with sharp edges and different solidification rates at each position; finally, under conditions of large undercooling, the entire alloy liquid advances in an arcuate direction. Additionally, it is observed that in Fig.4, the advancing position of the solidification interface during the solidification process is not fixed. Most start from the boundaries, some from both sides, and even from the center. This reflects the uncertainty of the solidification direction of the alloy liquid, attributed to the random nucleation positions of the alloy liquid. Throughout the undercooling experiment, the washing liquid solution contains more impurities, and the heterogeneous nucleation effect is evident. The alloy liquid tends to preferentially nucleate at the boundary between the washing liquid solution and the undercooled melt, leading to the above phenomena.

3.3 Grain refinement analysis

3.3.1 Analysis of grain refinement microstructure at low undercoolings

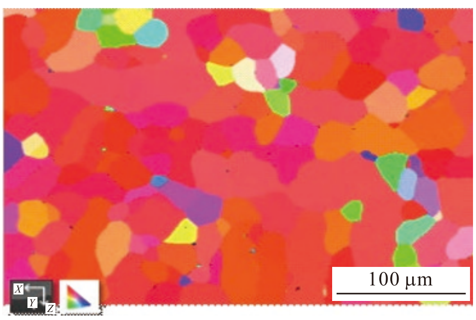


Fig.5 EBSD analysis of Ni65Cu33Co2 alloy with 74 K undercooling

Fig.5 shows the microstructure analysis results of Ni65Cu33Co2 alloy under undercooled conditions obtained through EBSD technology at 74K. EBSD analysis was performed on two alloy components, and it can be observed that there are many grains with similar orientations in their microstructures. Grains of the same or similar color indicate that they have the same orientation. Clearly, there are many grains with similar orientations. In the polar coordinate plot, we can also observe high-intensity textures with main orientations. No twinning phenomenon appeared in these textures. According to the definition of grain boundary misorientation, those with misorientation angles less than 5° are called low-angle grain boundaries, and those with angles greater than 15° are called high-angle grain boundaries. The proportion of low-angle grain boundaries is relatively high, while the proportion of high-angle grain boundaries is relatively low. In addition, the distribution of low-angle grain boundaries is also more concentrated. In conclusion, Fig.5 reveals the microstructural characteristics of the Ni65Cu33Co2 alloy under undercooled conditions through EBSD analysis, providing important references for further research.

3.3.2 Analysis of grain refinement microstructure at high undercoolings

Fig.6 illustrates the microstructure of Ni65Cu33Co2 alloy at 292 K under undercooled conditions as observed through EBSD. In the diffraction quality map 18a, it is evident that the alloy's structure exhibits more pronounced and linear polygonal grain boundaries under high undercooling conditions, resulting in smaller grain sizes at the same scale. This can be observed in the grain boundary map in Fig.6(b), where the proportion of high angle grain boundaries in the alloy's structure has significantly increased compared to conditions with lower undercooling (the grain boundary colors in the figure correspond to those in the EBSD map under small undercooling conditions). Furthermore, the grain boundary orientation difference map in Fig.6(d) reveals that the proportion of high angle grain boundaries exceeds 85%. Additionally, twin boundaries are visible in Fig.6(b), marking a notable difference from the structure under small undercooling conditions. The alloy's grain boundary orientation difference in Fig.6(c) displays a range of differently colored grains, indicating the random distribution of grain orientations in space. The corresponding pole in Fig.6(e) lacks any other high-intensity patterns, suggesting a departure from the refined structure under small undercooling conditions and indicating that dendritic remelting is not the primary factor contributing to the refinement of the structure under high undercooling conditions.

The use of the recrystallization theory to explain the refining mechanism at high undercoolings has become more reasonable in recent years. This is due to the accumulation of stress in dendrites during rapid solidification, which leads to plastic deformation and fracture of dendrites when the undercooling value exceeds a critical value. The plastic strain of the alloy structure provides power for recrystallization in the later recalescence period. In the case of Ni65Cu33Co2 alloy, grain refinement at high undercoolings results in a microstructure with flat grain boundaries, regular random orientation, and polygonal shapes in the majority of grains. Under high undercooling conditions, there is a significantly higher number of high angle grain boundaries compared to small undercooling conditions, and annealing twins also appear in the structure. These features indicate the occurrence of recrystallization at high undercoolings. The inter-engulfment between non-deformed grains causes the grain boundaries to change from curved to straight, resulting in the formation of high angle grain boundaries. During the recrystallization and grain growth process, only a few new textures are introduced, leading to an insignificant change in the main orientation of the texture and a decrease in observed pole Fig.intensity. TEM characterization analysis and bright field images of different regions of the Ni65Cu33Co2 alloy at high undercooling conditions reveal a large number of dislocations. These dislocations are a result of the superimposed stress of plastic deformation and dendritic crystal, providing a thermodynamic driving force for grain refinement and supporting the recrystallization mechanism. Overall, the abundance of dislocations in the alloy at high undercooling conditions provides evidence that the recrystallization mechanism is at play in the grain refinement process.

4 Conclusions

By using deep undercooling technology, we have

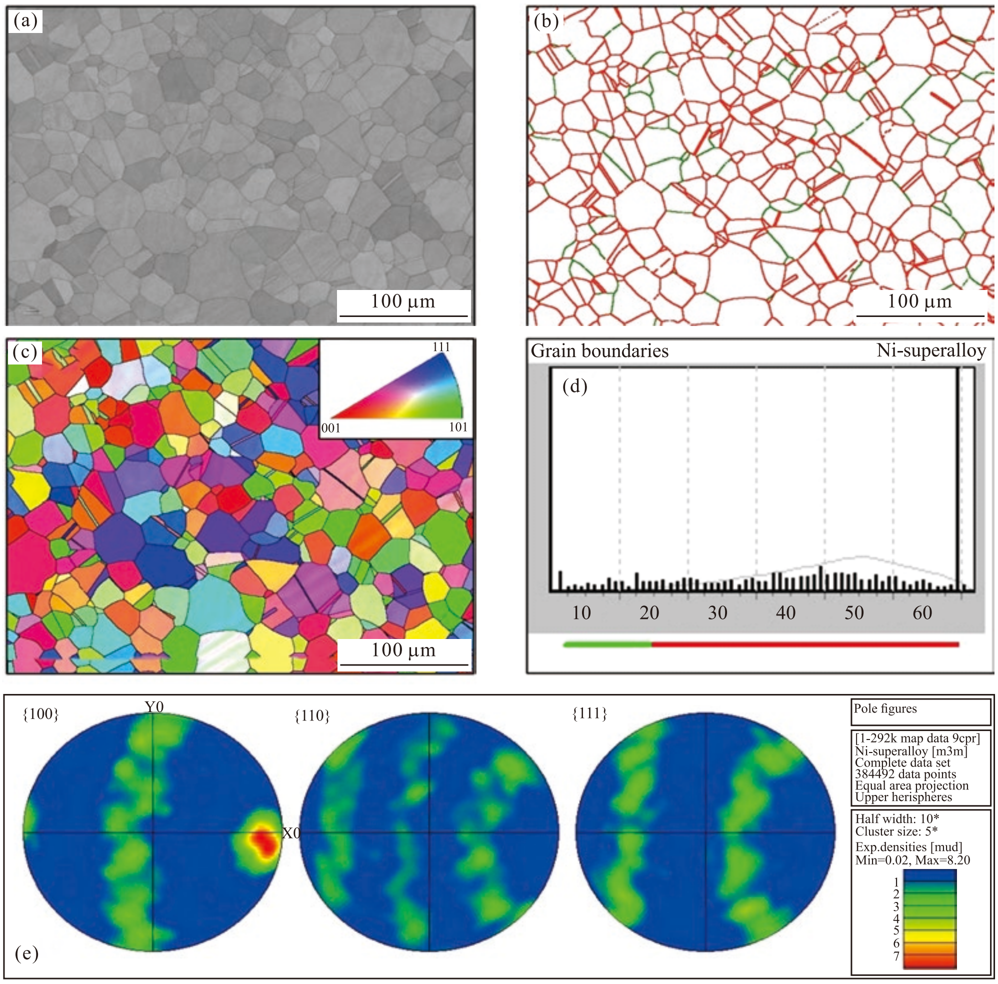


Fig.6 EBSD analyzed the Ni-Cu-Co alloy with the undercooling 292 K: (a) Diffraction quality map, (b) Large angle and small angle grain boundary map, (c) Crystal orientation map, (d) Grain boundary orientation difference distribution map, and (e) Pole figure

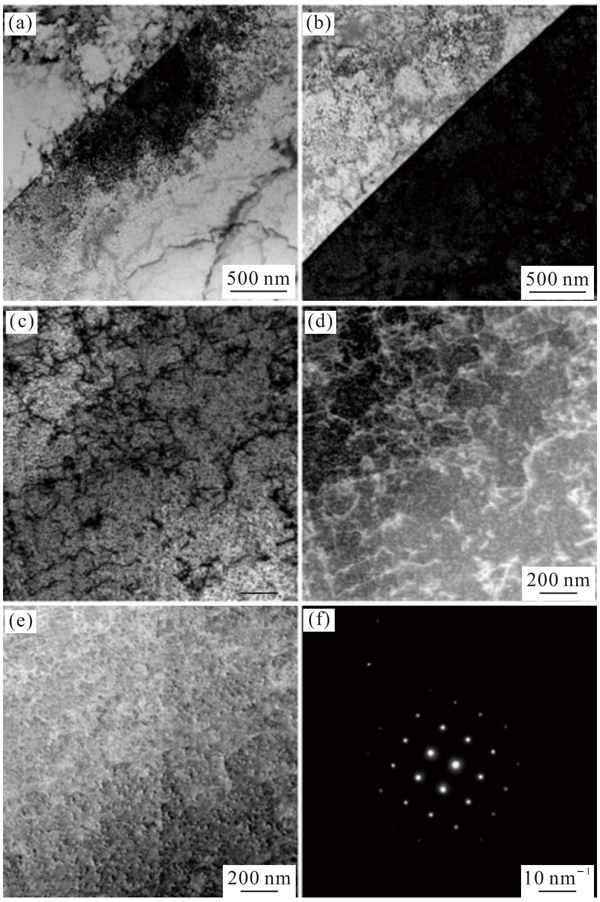


Fig.7 TEM bright field images of dislocations

studied the fine structure, solidification rate, solute distribution composition of Ni based ternary alloy melts and have made some important discoveries and conclusions. First, we successfully achieved high undercooling and nonequilibrium solidification of Ni based ternary alloy alloy, and obtained an undercooled structure with about 300 K. This provides an important basis for further exploration of the characteristics and applications of this alloy. Secondly, under deep undercooling conditions, we found that the two refinement mechanisms of Ni based ternary alloy alloy are consistent with the binary alloy (Ni-Cu). Specifically, at small undercooling conditions, grain refinement is due to dendrite remelting, while under large undercooling conditions, grain refinement is attributed to recrystallization, driven by stress and strain energy due to rapid solidification. These findings provide important clues for a deeper understanding of the mechanism in the alloy solidification. We also found that the addition of Co significantly improves the reheating effect and solidification rate of the melt, and enhances the stress effect and dendrite remelting leading to grain refinement. At the same time, the undercoolings of the alloy have been greatly improved. Finally, it is worth noting that the addition of Co is also beneficial for forming a non-discriminatory structure with a uniform element distribution under deep undercooling conditions, providing new ideas and methods for further optimizing the alloy preparation process. In conclusion, through this study, we have conducted a thorough discussion on the deep undercooling characteristics and microstructure of Ni based ternary alloy ternary alloy, and have achieved some exciting results, which will provide important references and guidance for the design and preparation of alloys.

Conflict of interest

All authors declare that there are no competing interests.

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