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Microstructures and mechanical properties of directionally solidified multi-element Nb-Si alloy

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Abstract: Nb-Si system in-situ composites with nominal composition of Nb-21Ti-16Si-7Cr-3Al-2Hf (mole fraction, %) were prepared by directional solidification technology in an optical floating zone furnace at the growth rates of 3, 5, 8 and 12 mm/h. The microstructures and mechanical properties were investigated as a function of the growth rate. The results show that the solidification microstructures consist of Nb solid solution (Nbss), α-Nb_5Si_3 and β-Nb_5Si_3. Silicides with high aspect ratio grow continuously and straightly when the growth rate is 5mm/h; lower or higher growth rates are disadvantageous for the formation of favorable directional microstructures. The compressive strength at 1 250 °C and room-temperature fracture toughness of the specimen with the growth rate of 5 mm/h that exhibits excellent mechanical properties are 415 MPa and 22.4 MPa·m$^{1/2}$, respectively.
Key words: Nb-Si system in-situ composite; directional solidification; microstructure; mechanical property

1 Introduction

The development of aircraft industry demands higher thrust mass ratio and fuel efficiency of jet engine, which is strongly dependent on the inlet temperature of high-pressure turbine (HPT). Metal surface temperatures of the hottest sections on the advanced turbine engine blades approach about 1 150 ºC, which is essentially the limit for the most advanced single-crystal Ni-based superalloys[1]. In order to increase the temperature capability of airfoils further, new material systems have to be exploited. In recent years, Nb-silicide in-situ composites, which have received substantial attention because of their high melting point (>1 750 °C), low density (6.6−7.2 g/cm^3, as compared with about 9.2 g/cm^3 for Ni-based superalloys), good balance between high temperature strength and low temperature damage tolerance, show considerable potential for the application in the field of ultrahigh temperature structural materials[2-4]. However, low room temperature fracture toughness is a serious obstacle to the practical applications of the composites. A great deal of effort has been made to balance high temperature strength and room temperature toughness by adjusting fabrication technology and controlling microstructures. Previous work show that among the various preparation techniques, such as arc-melting[5−7], sputter deposition[8], powder sintering[9−15] and laser forming[16], directional solidification is a useful approach to modify the microstructures and improve the mechanical properties of Nb-silicide in situ composites[17−20]. Therefore, Nb-Si alloys are prepared by directional solidification in an optical floating zone (OFZ) furnace in the present study. The effect of growth rates on the microstructures and mechanical properties was investigated.

2 Experimental

The master alloy ingot with nominal composition of Nb-21Ti-16Si-7Cr-3Al-2Hf was prepared by consumable arc-melting method. The raw materials were high pure Nb, Ti, Si, Cr, Al, and Hf (Nb 99.8%, Ti, Cr, Al, Hf: 99.9%, Si: 99.999%, mass fraction). In order to ensure chemical homogeneity, the alloy ingot was re-melted five times. The dimensions of the master alloy ingot were about 60 mm in diameter and 150 mm in length. The rods used for directional solidification with dimensions of 9 mm in diameter and 70 mm in length

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were cut from the master alloy ingot by electro-discharge machining (EDM) and each surface was mechanically ground. The directional solidification experiments were carried out in an FZ-35WHV optical floating zone (OFZ) melting furnace under an Ar gas flowing atmosphere at the growth rates of 3, 5, 8 and 12mm/h. To facilitate description in the paper, the alloys directionally solidified at 3, 5, 8, 12 mm/h are referred to as DS3, DS5, DS8 and DS12 alloy, respectively.

The microstructural analysis of the alloys was performed by scanning electron microscopy (SEM), energy dispersive X-ray spectroscopy (EDS) and X-ray diffractometry (XRD). The mechanical properties were evaluated by high temperature compression tests and room temperature fracture toughness measurements. The compression specimens with the size of d 6 mm × 9 mm were cut from the directional solidification rods by EDM and the loading direction was parallel to the solidification direction. The compression tests were conducted using a Gleeble testing machine at 1 250 ℃ in vacuum with a nominal strain rate of $5×10^{-3}$ s^{-1}. The room temperature fracture toughness was determined by three-point bending tests using single notched specimens with the dimension of 3 mm × 6 mm × 30 mm. A single notch perpendicular to the solidification direction with about 3 mm in length was introduced at half of the length by EDM.

3 Results and discussion

3.1 Constituent phases and microstructures

Figure 1 shows the XRD pattern of the master alloy and directionally solidified alloys. It can be seen from Fig.1 that the master alloy consists of three phases of Nb solid solution (denoted as Nbss for short), α-Nb$_5$Si$_3$ and β-Nb$_5$Si$_3$. After directional solidification at different rates, constituent phases of the alloys do not change as those of the master alloy. The typical microstructures of master alloy and directionally solidified alloys are shown in Fig.2. Combining the results of XRD and EDS analysis, the bright phase and grey phase in Fig.2 are Nbss and Nb$_5$Si$_3$, respectively. It is difficult to differentiate α-Nb$_5$Si$_3$ from β-Nb$_5$Si$_3$ by contrast of back-scattered electron images (BEI). Fig.2(a) reveals that besides bright dendritic Nbss and large gray Nb$_5$Si$_3$ block, there are massive fine rosette. Fig.2(b)−(e) display the transversal section microstructures of directionally solidified alloys. Silicides in DS3 alloy (Fig.2(b)) distribute in the Nbss matrix with network and block structure. Transversal microstructure of DS12 alloy is similar to that of DS3 alloy; however, silicide size of DS12 alloy is smaller than that of DS3 alloy. Fig.2(c) and (d) show that except a few of big size silicide phases, the rest of silicides are extremely fine. Fig.2(f)−(i)

illustrate the longitudinal section microstructures of directionally solidified alloys. The microstructures of DS3 alloy and DS5 alloy are characterized by lamellar silicides aligned parallel to the growth direction and embedded within the Nbss matrix, indicating that coupled growth of Nbss and Nb$_5$Si$_3$ is obtained at 3mm/h and 5mm/h. Some growth defects, such as discontinuous silicides and transversal dendrites, are also observed. For DS8 alloy (Fig.2(h)), discontinuous and transversal faults are more significant than DS3 and DS5 alloy although part of silicides grow along longitudinal direction. When the growth rate increases to 12mm/h, silicides are no preferential orientation and directional growth characters completely disappear, as shown in Fig.2(i).
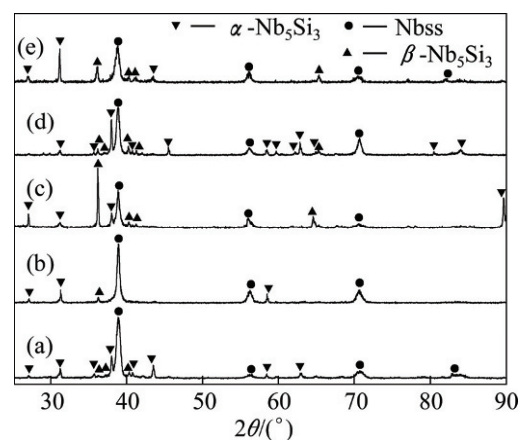

Fig.1 XRD patterns of master alloy and directionally solidified alloys with different growth rates: (a) Master alloy; (b) 3 mm/h; (c) 5 mm/h; (d) 8 mm/h; (e) 12 mm/h

Based on the above-mentioned results, it can be concluded that the longitudinal continuous growth of silicides is destroyed gradually with increasing growth rate. Low growth rate is beneficial to coupled growth, but if the growth rate is too low, such as 3 mm/h, because of low temperature gradient the growth of silicides along transversal direction is developed, resulting in coarse silicides. Thus, it is only appropriate growth rate, such as 5 mm/h, that realizes coupled growth of Nbss and silicide and formation of silicides with high aspect ratio simultaneously.

3.2 High temperature compressive properties

Figure 3 shows compressive stress-strain curves of master alloy and directionally solidified alloys at 1 250 ℃. As seen in Fig.3, the values of compressive strength for DS3 alloy and DS5 alloy are 445 MPa and 415 MPa, respectively, which are about 36.5% and 27.3% higher than that of master alloy (326 MPa). The improvement of compressive strength at high temperature for DS3 alloy and DS5 alloy may be attributed to the formation of the columnar structure oriented in the growth direction. The
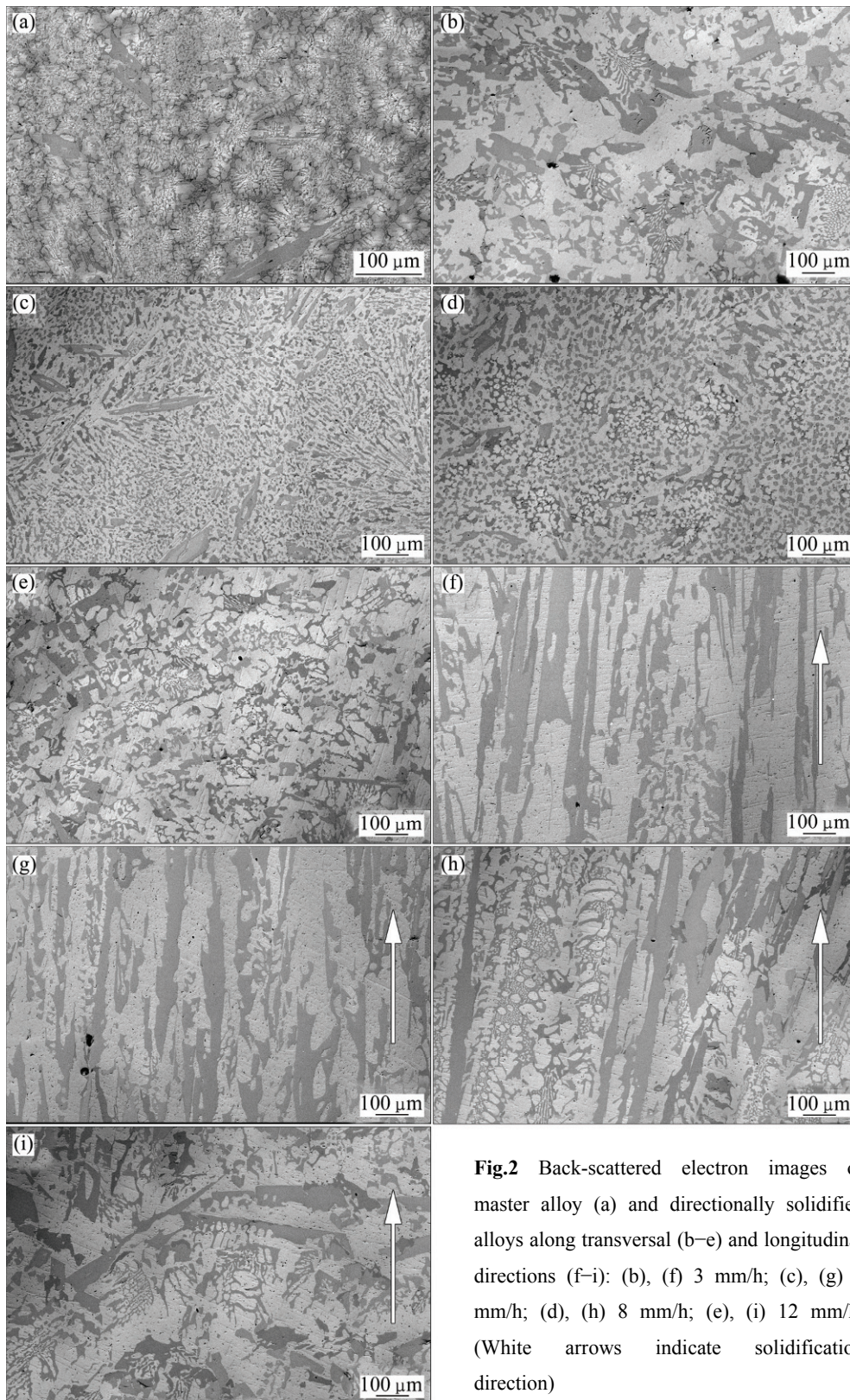
Fig.2 Back-scattered electron images of master alloy (a) and directionally solidified alloys along transversal (b−e) and longitudinal directions (f−i): (b), (f) 3 mm/h; (c), (g) 5 mm/h; (d), (h) 8 mm/h; (e), (i) 12 mm/h. (White arrows indicate solidification direction)

columnar structure is much more difficult to deform than the equiaxed structure. Moreover, transversal interfaces between Nbss and Nb_5Si_3 are weak and easy to slide and break under circumstances of elevated temperature and longitudinal stress. Unlike interfaces distributing at random in master alloy, transversal interfaces in DS3 alloy and DS5 alloy are few and most interfaces are parallel to the loading direction, which requires higher

stress to achieve interfacial sliding and breaking. Increasing the growth rate to 8 mm/h and 12 mm/h, the compressive strength decreases due to a reduction of longitudinal continuous growth tendency of silicides.

Figure 4 reveals the microstructures of master alloy and DS3 alloy after high temperature compression tests.
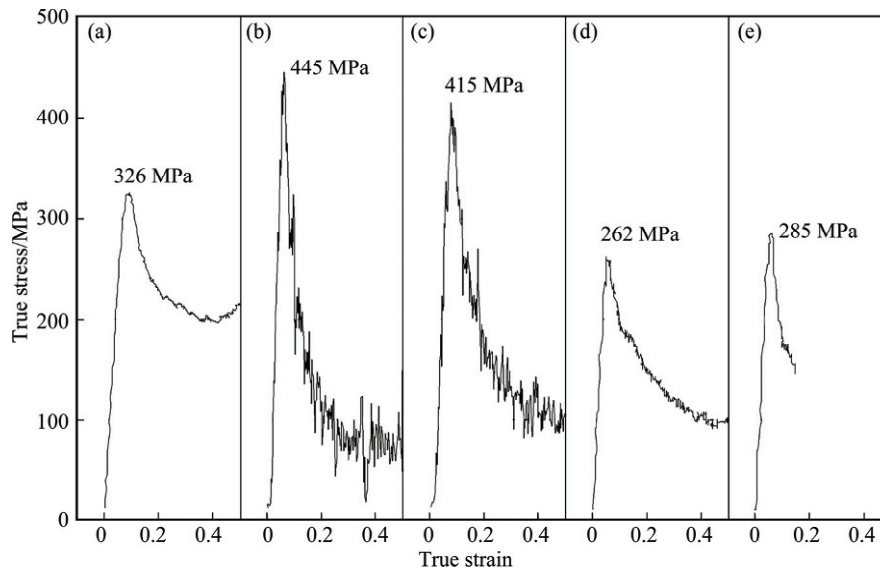

Fig.3 Compressive stress—strain curves of master alloy and directionally solidified alloys at 1 250 °C: (a) Master alloy; (b) 3 mm/h; (c) 5 mm/h; (d) 8 mm/h; (e) 12 mm/h;
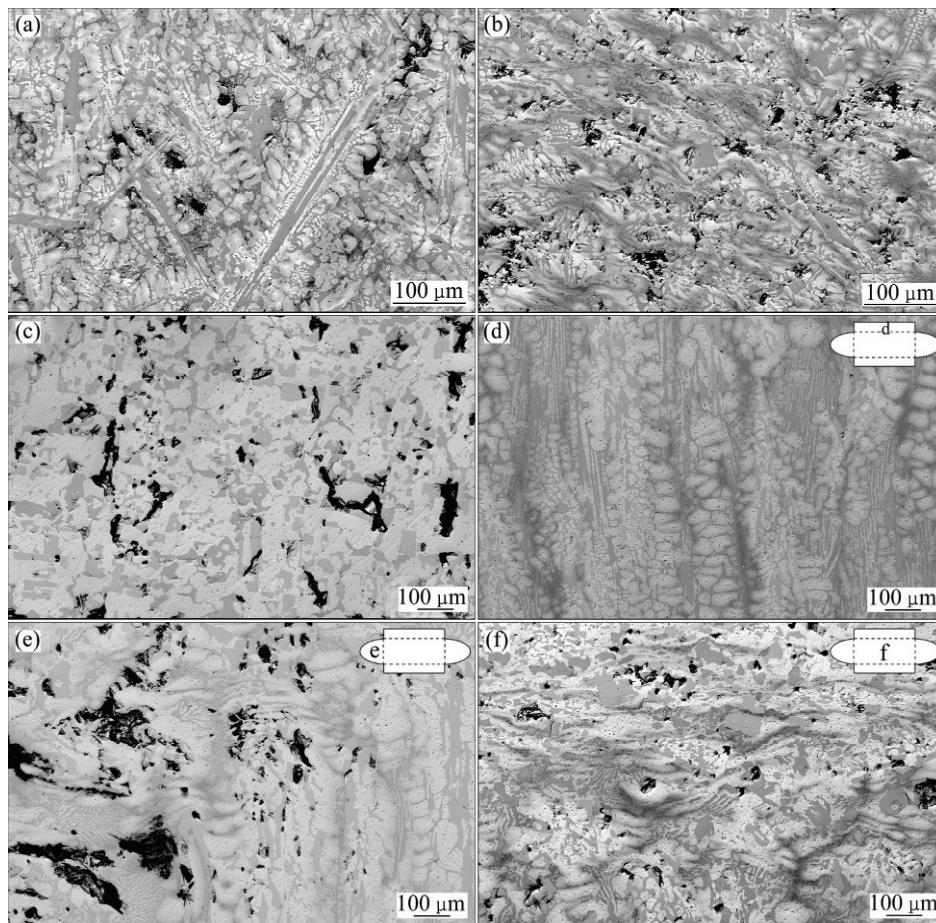

Fig.4 Microstructures of master alloy (a, b) and DS3 alloy (c−f) after high temperature compression tests (Insets at top right corner of (d), (e), (f) illustrate locations for microstructural observation): (a, c) Perpendicular to loading direction; (b, d−f) Parallel to loading direction; (d) At top of specimen; (f) At side of specimen; (e) In middle of specimen

As shown in Fig.4(a) and 4(b), whether from transversal direction or longitudinal direction, silicides in master alloy cracked and some holes are formed after compression at 1 250 °C. Obvious distortion parallel to loading direction is observed in Fig.4(b). For DS3 alloy, holes also appear in the microstructure perpendicular to loading direction. However, microstructural differences at different locations of the specimen along loading direction are evident. At the top of the specimen, except slight distortion, silicides remain longitudinal alignment morphology basically and no cracks or holes are present, as shown in Fig.4(d). At the side of the specimen compressed to drum shape (Fig.4(e)), silicides deform seriously and lose characters of unidirectional arrangement. Furthermore, big holes emerge after compression. In the middle of the specimen (Fig.4(f)), the morphology is similar to that of the side except the hole size is smaller.

3.3 Room temperature fracture toughness

Figure 5 shows the results of room temperature fracture toughness (K_Q) test of master alloy and directionally solidified alloys with different growth rates. The room temperature fracture toughness increases with increasing growth rate when the rate is lower than 5 mm/h. Nevertheless, as the growth rate increases further, K_Q decreases visibly. Compared with the master alloy, the K_Q values of DS3 alloy and DS5 alloy are 17.99 MPa·m$^{1/2}$ and 22.4 MPa·m$^{1/2}$, respectively, indicating increase of 27.4% and 58.6%. These improvements are attributed to the favorable directional microstructures. As Fig.6 shows, during the cracks propagation along the direction perpendicular to the solidification direction, the crack-tip energy is partly absorbed and the alloys are toughened accordingly due to crack deflections and microcracks locating at the interfaces between Nbss and

Nb$_5$Si$_3$. Toughening mechanism of crack deflections and microcracks is weakened for DS8 alloy and DS12 alloy because of their less or even not oriented silicides. Besides fracture toughness, Fig.5 also shows displacement at the maximum load. The change regularity of displacement is similar to that of K_Q. Higher displacement means better plasticity. Thus, DS5 alloy exhibits the best plasticity at room temperature.

Room temperature fracture toughness of DS5 alloy is higher than that of DS3 alloy because silicides in the former possess higher aspect ratio. Similar volume of silicides for DS3 alloy and DS5 alloy are obtained due to their same composition. Consequently, silicide distribution in Nbss matrix is much closer for DS5 alloy with higher aspect ratio, which increases the opportunities of crack deflection and microcrack at the interfaces between Nbss and Nb$_5$Si$_3$ (Fig.7), contributing to more crack-tip energy being absorbed and higher fracture toughness.
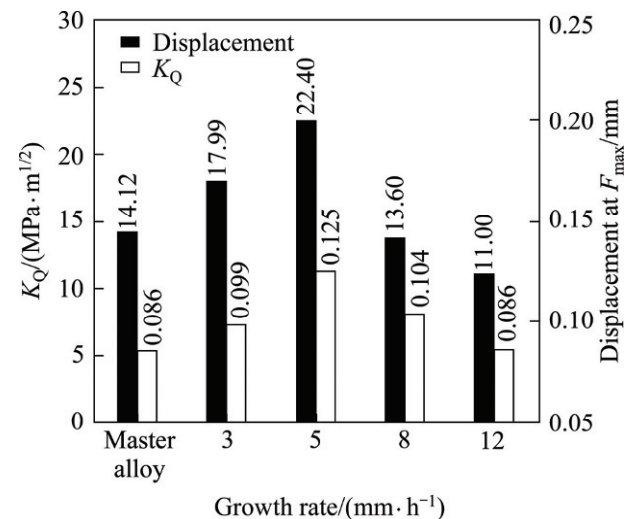

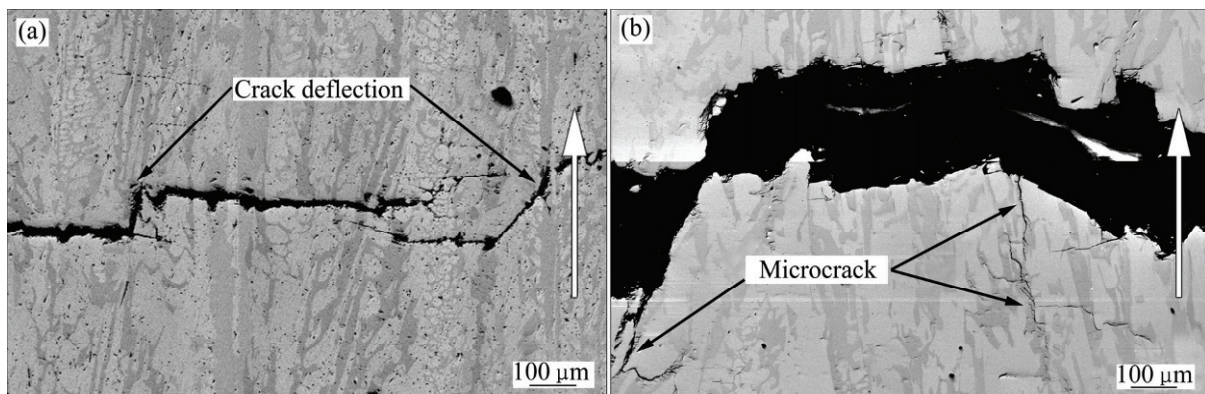**Fig.5** Three-point bending test results of master alloy and directionally solidified alloys with different growth rates

Fig.6 Crack propagation behavior of directionally solidified alloys with different growth rates: (a) 3 mm/h; (b) 5 mm/h
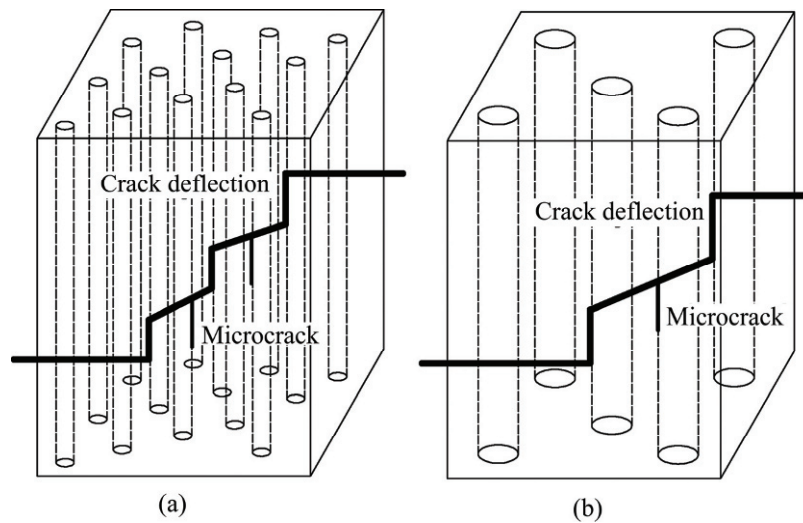
Fig.7 Schematic diagram of crack propagation behavior in the alloys with different aspect ratios: (a) High aspect ratio; (b) Low aspect ratio

4 Conclusions

1) Nb-silicides in-situ composites prepared by directional solidification consist of Nbss, α-Nb$_5$Si$_3$ and β-Nb$_5$Si$_3$ phases. The microstructures of directionally solidified alloys strongly depend on the growth rate. Higher growth rate leads to non-oriented silicides, but lower growth rate makes silicide transversal size coarse. The growth rate of 5mm/h is advantageous for coupled growth and formation of silicides with a high aspect ratio.

2) High temperature compressive strength and room temperature fracture toughness of DS3 alloy and DS5 alloy are higher than those of the master alloy. The enhancement in compressive strength is due to the formation of columnar structure and considerable interfaces which are parallel with the loading direction. An improvement of the fracture toughness is as a result of crack deflections and microcracks occurred at interfaces between Nbss and Nb$_5$Si$_3$. The fracture toughness of DS5 alloy at room temperature is higher than that of DS3 alloy, which is attributed to silicides with a higher aspect ratio.

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