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# Environmental effect on mechanical properties of recrystallized $L1_2$-type $Ni_3(Si, Ti)$ intermetallics

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The environmental effect on the mechanical properties of boron-doped and undoped $Ni_3(Si, Ti)$ polycrystals was investigated by tensile testing in air from room temperature to 1073 K, and the results were compared with those obtained previously by tensile testing in vacuum. The environmental effect for the $Ni_3(Si, Ti)$ alloys was significant at ambient temperatures whereas that for the boron-doped $Ni_3(Si, Ti)$ alloys was considerable at elevated temperatures. When these samples at associated temperatures were tensile tested in air and also at low strain rate, intergranular fracture was dominant. It was suggested that the environmental embrittlements at low and high temperatures were due to hydrogen and oxygen absorbed from the air, respectively, and were caused by the weakening of the grain-boundary cohesion. It was proposed that boron competing with hydrogen, for site occupation or for its effectiveness at grain boundaries, has the effect of suppressing hydrogen embrittlement, whereas it was suggested that the low-melting phases, consisting of boron and oxygen (and/or constituent atoms), may be responsible for the ductility loss in the boron-doped $Ni_3(Si, Ti)$ alloys.

## 1. Introduction

By the development of alloying techniques and material processing for intermetallic compounds, some fabricable intermetallic compounds have been produced, an example being the $L1_2$-type $Ni_3(Si, Ti)$ alloys which have been studied by the present authors [1, 2]. However, as ductile intermetallic compounds are documented, it has been recognized recently that the mechanical properties of these alloys are extremely influenced by the test environment. The tensile ductility and the associated fracturing strongly depended upon the test environment, with the worst ductility in cathodic charging of hydrogen or hydrogen-gas exposure, and with lower ductilities observed in air rather than in vacuum. Also, there was a strong effect of strain rate on the ductility, with lower ductilities observed at a lower rather than at higher strain rates. These embrittlements are commonly accompanied by an intergranular crack path postulated to be associated with hydrogen atoms. Indeed, the $Ni_3(Si, Ti)$ alloys showed the environmental effect on the tensile elongation at ambient temperatures [1]. This kind of environmental effect has been reviewed in several articles [3-5].

Recently, different kinds of environmental effects have been reported. Oxygen, absorbed from the air during high-temperature testing, has been shown to embrittle the $Ni_3Al$-based $L1_2$ compounds rendered ductile by the addition of boron [6]. The tensile ductilities measured at elevated temperatures were found to be a strong function of the test environment, whether in air or in vacuum; the samples tensile-tested in air showed a severe loss of ductility. This environmental embrittlement also depended upon strain rate and produced grain-boundary fracture paths, thus being phenomenologically very similar to the hydrogen-related embrittlement, although the operative temperature is different. However, data for other intermetallic compounds remain scarce.

In the preceding work [2], the mechanical properties of the $Ni_3(Si, Ti)$ alloys were observed as a function of the test temperature and with respect to the doping of a small amount of boron. These data were taken from the tensile tests in vacuum, except for data at 77 K. In the present work, the same materials as those tested in the previous work (in terms of alloy composition and microstructure) were tensile tested in air, and then compared with the previous results. The tensile tests were performed at temperatures from room temperature to 1173 K. Also, the strain-rate effect on the mechanical properties of these alloys were investigated, particularly at high temperatures where the embrittlement due to gaseous oxygen in air appeared. These tensile data were also correlated with fractographic observation.

## 2. Experimental procedure

The chemical compositions of the alloys used in this work were given in Table I of the preceding paper [2]: $Ni_{79.5}Si_{11}Ti_{9.5}$ and $Ni_{79.5}Si_{11}Ti_{9.5}$ + 50 p.p.m. B (denoted $Ni_3(Si, Ti)$ alloy and boron-doped $Ni_3(Si, Ti)$ alloy, respectively).

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The alloy preparation, heat-treatment (for homogenization and recrystallization), tensile-specimen preparation and tensile testing were done using exactly the same procedures as those in the preceding work [2]. The tensile samples were heated within 1 h to the test temperature and then held for about 0.5 h at this temperature before pulling. Most of the mechanical tests were carried out at a nominal strain rate of 1.2 $\times 10^{-3}$ sec$^{-1}$. In order to determine the strain-rate effect on the mechanical properties of these alloys at high temperatures, a number of samples were tensile tested at various strain rates (from $10^{-4}$ to $10^{-1}$ sec$^{-1}$) in air and in vacuum. After tensile testing, the fracture surfaces of the samples were examined by scanning electron microscopy (SEM).

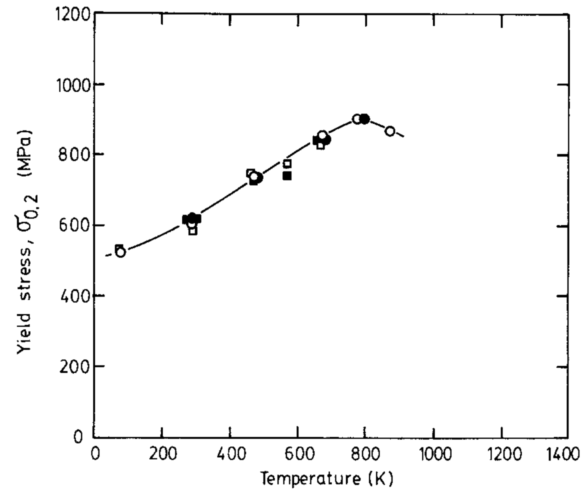

*Figure 1* Variations of the 0.2% yield stress of (□, ■) Ni$_3$(Si, Ti) alloys and (○, ●) boron-doped (50 p.p.m. B) Ni$_3$(Si, Ti) alloys with the test temperature. The tests were performed in (□; ○) vacuum and (■, ●) air.

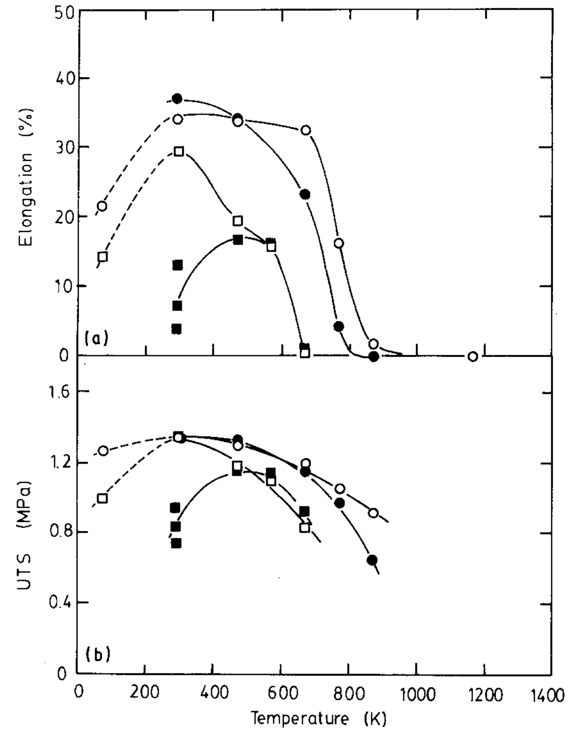

*Figure 2* Variations of (a) the elongation and (b) the ultimate tensile stress (UTS) of (□, ■) Ni$_3$(Si, Ti) alloys and (○, ●) boron-doped (50 p.p.m. B) Ni$_3$(Si, Ti) alloys, with test temperature. The results are compared between the two environmental conditions: (□, ○) vacuum, (■, ●) air.

## 3. Results

### 3.1. Variations of tensile properties with temperature

The yield stresses in two alloys, undoped and boron-doped, are shown in Fig. 1 as a function of the testing temperature, and compared with those [2] tested in vacuum. For both alloys, the yield stresses obtained by the test in air were basically identical to those obtained by the test in vacuum over whole test temperature range. Thus, it is obvious that the yield stress in the two alloys was little affected by the environmental medium.

Fig. 2 shows the variations of the elongation (Fig. 2a) and the ultimate tensile stress (UTS) (Fig. 2b) with testing temperature for the Ni$_3$(Si, Ti) alloys and the boron-doped Ni$_3$(Si, Ti) alloys, respectively. Each datum point was again compared between the two environmental media.

First, for the Ni$_3$(Si, Ti) alloys, the discrepancy in the elongation and the UTS values between the two environmental media was significant at ambient temperatures. At room temperature, the elongation value was about 29% when the sample was tested in vacuum, whereas they were about 4% to 13% when the samples were tested in air. Similarly, the UTS values were reduced from 1.35 GPa to about 0.75 to 0.95 GPa when the test environment changed from vacuum to air. Also, an important result seen in this figure is that as the test temperature increases the elongation of the samples tested in air increases, and at 473 K(200 °C) almost recovers to the values of the samples tested in vacuum. The UTS values also exhibited similar changes with increasing temperature; at 473 K the UTS values were almost identical in the two environmental conditions. At higher temperatures, above 473 K, a meaningful difference in the two properties was not recognized between the two environmental media, although apparent ductilities disappeared at temperatures above 673 K.

On the other hand, for the boron-doped Ni$_3$(Si, Ti) alloys, a quite different result was observed. At room temperature, the elongation and the UTS values of the boron-doped Ni$_3$(Si, Ti) alloys were almost the same in the two environmental media and were higher than those of the undoped Ni$_3$(Si, Ti) alloys. However, as the test temperature increases, the discrepancy of the elongation and the UTS between vacuum and air began to appear. This ductility loss was considerable at temperatures above 473 K.

Thus, it is concluded from this figure that the environmental embrittlement for the undoped Ni$_3$(Si, Ti) alloys is significant at ambient temperatures but not at elevated temperatures whereas that for the boron-doped Ni$_3$(Si, Ti) alloys is not evident at ambient temperatures but is so at elevated temperatures.

### 3.2. Strain-rate effect on the tensile properties at elevated temperatures

In investigating the environmental effect due to hydrogen which appears at ambient temperatures, the strain rate usually had a pronounced influence on the magnitude of its effect. A similar strain-rate effect might be observed in the environmental effect appearing at elevated temperatures. This experiment was performed at 573 K for the undoped $Ni_3$(Si, Ti) alloys and at 773 K for the boron-doped $Ni_3$(Si, Ti) alloys. (Each test temperature was chosen in order to achieve a certain amount of ductility loss due to high-temperature embrittlement caused by oxygen (see Fig. 2); in the former alloys, the hydrogen embrittlement prevails at low temperatures below 573 K, but apparent ductilities cannot be observed at higher temperatures, above 573 K. On the other hand, in the latter alloys, oxygen embrittlement does not occur at too low temperatures, below 773 K, but neither can apparent ductilities cannot be observed at too high temperatures, above 773 K.)

Fig. 3 summarizes the variations of the elongation (Fig. 3a), the UTS (Fig. 3b) and the yield stress (Fig. 3c) with the strain rate for the undoped $Ni_3$(Si, Ti) alloys. First, not only the yield stress but also the elongation and the UTS were basically independent of the testing environment, although the elongation showed a higher value in air than in vacuum at the highest strain rate tested. This result is consistent with the results of the undoped $Ni_3$(Si, Ti) alloys observed in Figs 1 and 2. A second point is that the yield stress was independent of the strain rate but the elongation and the UTS were very dependent on the strain rate; both values decreased with decreasing strain rate.

Fig. 4 summarizes the variations of the elongation (Fig. 4a), the UTS (Fig. 4b) and the yield stress (Fig. 4c) with the strain rate for the boron-doped $Ni_3$(Si, Ti) alloys. Fig. 4c clearly shows that the yield stress was independent of both strain rate and testing environment, whereas both tensile elongation and UTS are strongly dependent on these two factors; two values decreased with decreasing strain rate and also were lower in air than in vacuum. These results were also consistent with the results of the boron-doped $Ni_3$(Si, Ti) alloys shown in Figs 1 and 2.

### 3.3. Fracture behaviour

In the preceding paper [2], the fractographs of the undoped and boron-doped $Ni_3$(Si, Ti) alloys which were tested in vacuum were shown as a function of the testing temperature. Descriptions were given in terms of the temperature dependence of the fracture patterns. It was found in the correlation between fractographs and ductility, that as the elongation value (or UTS) increased, the fracture pattern changed from the intergranular to the transgranular pattern [2].

First, for the undoped $Ni_3$(Si, Ti) alloys, the difference in the fractographs between the two environmental media was significant at ambient temperatures. Fig. 5 shows the fracture patterns of the samples which were tensile tested at room temperature in vacuum (Fig. 5a) and in air (Fig. 5b). It is clearly seen that the

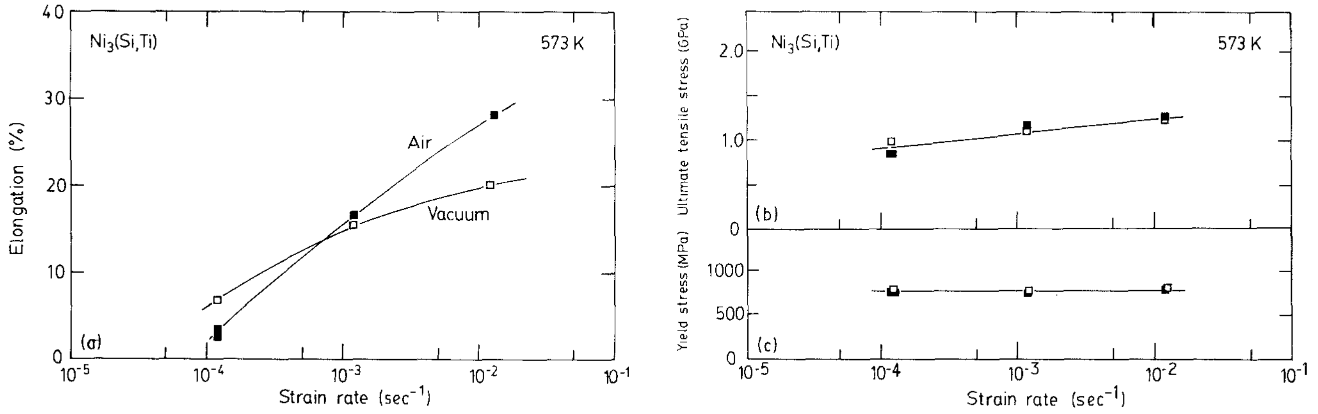

*Figure 3* Variations of (a) the elongation, (b) the UTS and (c) the yield stress of the $Ni_3$(Si, Ti) alloys with strain rate.

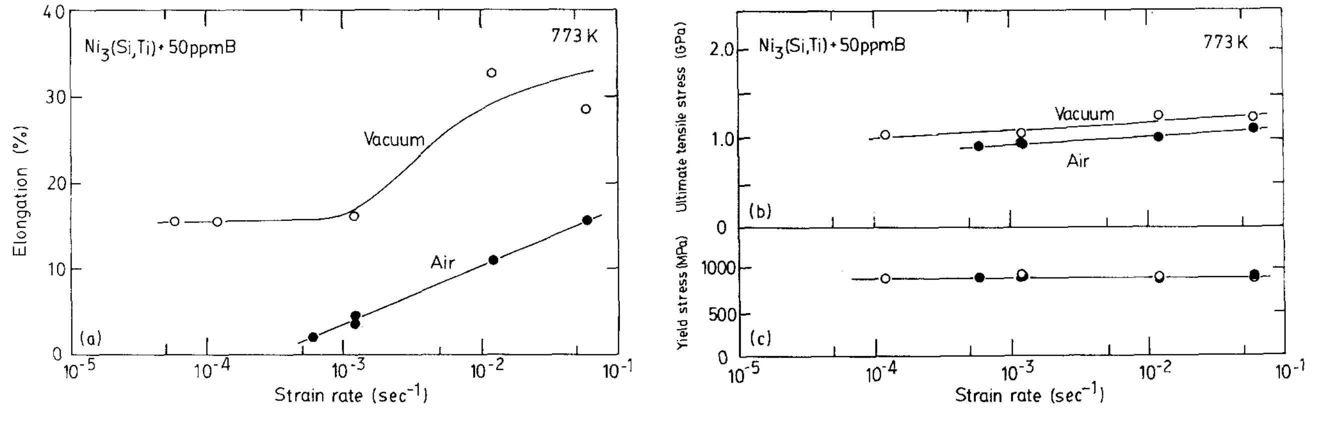

*Figure 4* Variations of (a) the elongation, (b) the UTS and (c) the yield stress of boron-doped $Ni_3$(Si, Ti) alloys with strain rate.

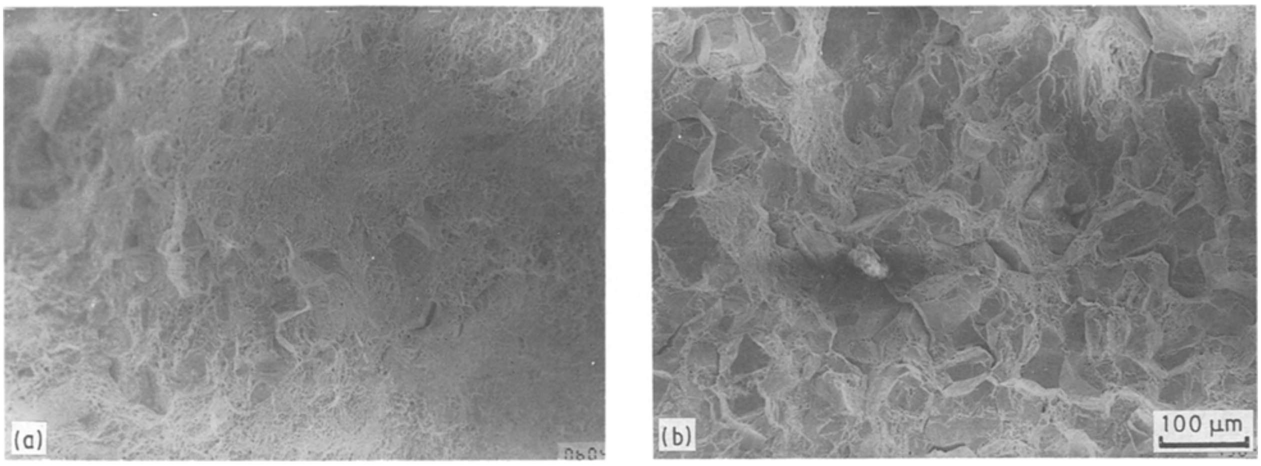

*Figure 5* Fracture patterns of the $Ni_3(Si, Ti)$ alloys which were tensile tested at room temperature (a) in vacuum and (b) in air.

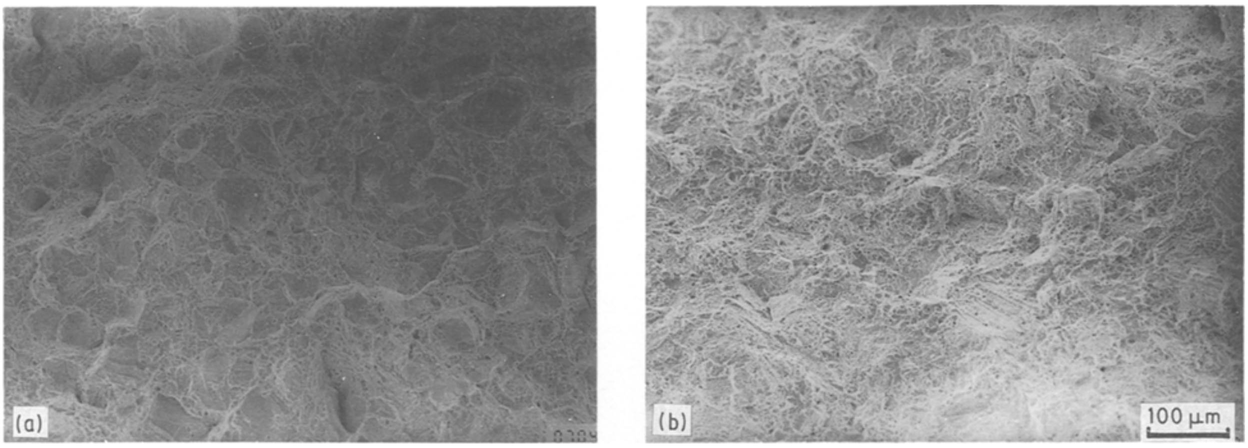

*Figure 6* Fracture patterns of the $Ni_3(Si, Ti)$ alloys which were tensile tested at 573 K (a) in vacuum and (b) in air.

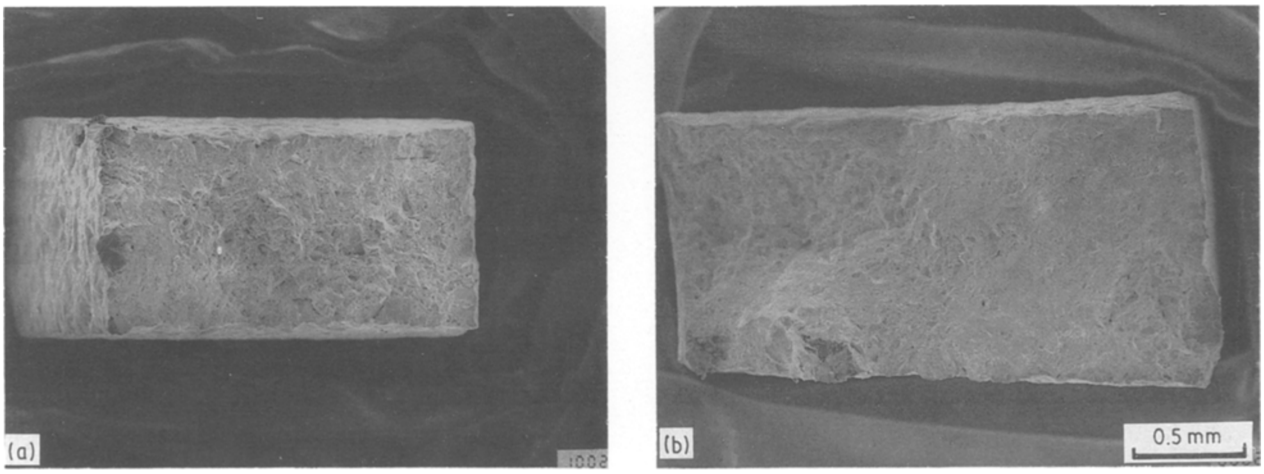

*Figure 7* Fracture patterns of the boron-doped $Ni_3(Si, Ti)$ alloys which were tensile tested at room temperature (a) in vacuum and (b) in air.

sample tested in vacuum showed ductile transgranular fracture patterns mixed with a small amount of intergranular fracture patterns, whereas the sample tested in air showed a mixture of transgranular and intergranular fracture patterns, thus corresponding to the fact that the former sample was more ductile than the latter, as shown in Fig. 2. On the other hand, Fig. 6 shows the fracture patterns of the samples which were tensile tested at 573 K in vacuum (Fig. 6a) and in air (Fig. 6b). These two fracture patterns are quite similar, thus corresponding to the result that two samples showed almost the same elongation and the UTS values as shown in Fig. 2.

Next, for the boron-doped $Ni_3(Si, Ti)$ alloys, the difference in fractography between the two environmental media was significant at elevated temperatures. Fig. 7 shows the fracture patterns of the samples which were tensile tested at room temperature in vacuum (Fig. 7a) and in air (Fig. 7b). These two fracture patterns are quite similar and showed mostly ductile

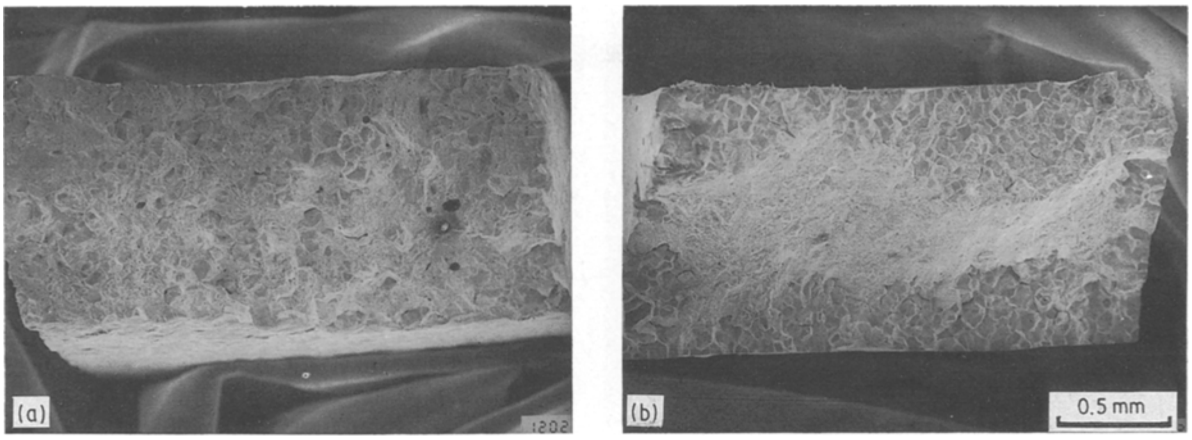

*Figure 8* Fracture patterns of the boron-doped $Ni_3(Si, Ti)$ alloys which were tensile tested at 773 K (a) in vacuum and (b) in air.

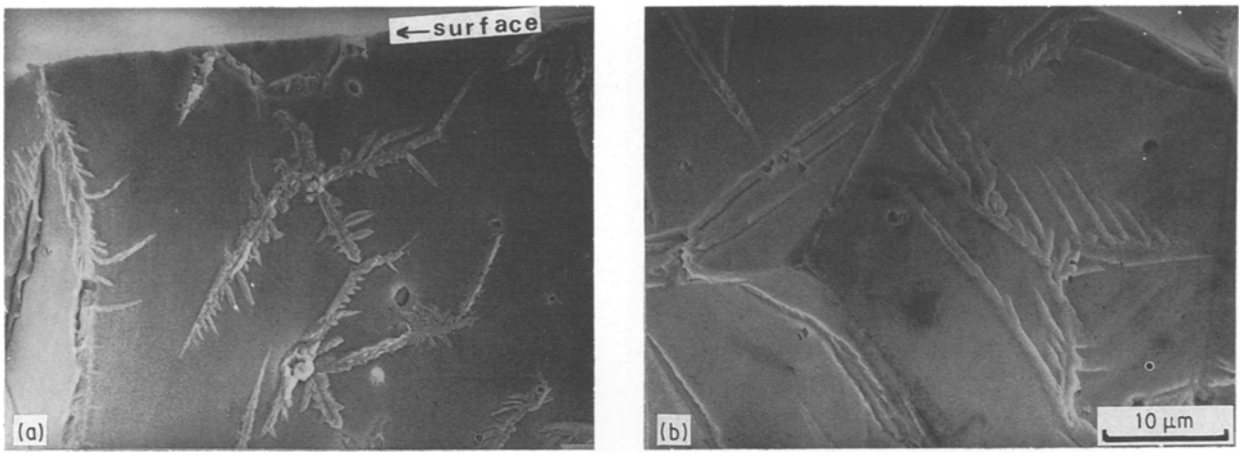

*Figure 9* Highly magnified photographs taken at the grain-boundary facets appearing near the free surface of the boron-doped $Ni_3(Si, Ti)$ alloy which was tensile tested at 773 K in air.

transgranular fracture patterns. Thus, this result corresponds well with the result that two samples showed almost identical and high values of elongation and UTS, as shown in Fig. 2. On the other hand, Fig. 8 shows the fracture patterns of the samples which were tensile tested at 773 K in vacuum (Fig. 8a) and in air (Fig. 8b). Here, very interesting fracture patterns were observed. The sample tested in vacuum exhibited a mixture of intergranular and transgranular fracture patterns throughout the whole cross-section of the fractured surface (Fig. 8a). However, the sample tested in air showed almost all intergranular fracture patterns in the region near to the sample's free surface and the transgranular fracture patterns tended to be more dominant on approaching the sample's interior (Fig. 8b). Thus, it appears that the intergranular fracture which appeared near the sample surface was caused by testing in the air medium.

More detailed fractographic observations were made on the boron-doped $Ni_3(Si, Ti)$ alloys which were tensile tested in air at 773 K. Fig. 9 shows the highly magnified photographs taken at the grain-boundary facets appearing near the free surface of this sample. A number of peculiar patterns, i.e. "dendritic" microstructures, were observed on the grain-boundary planes or grain-boundary corners. This kind of pattern was only observed in samples which were boron-doped, tested in air and at elevated temperatures. Although structural and chemical analyses were not performed on these dendritic microstructures, this morphology suggests that this phase might be a low-melting phase created during loading in air and at high temperature.

The surface-affected zone covered by the intergranular fracture in the boron-doped $Ni_3(Si, Ti)$ alloys

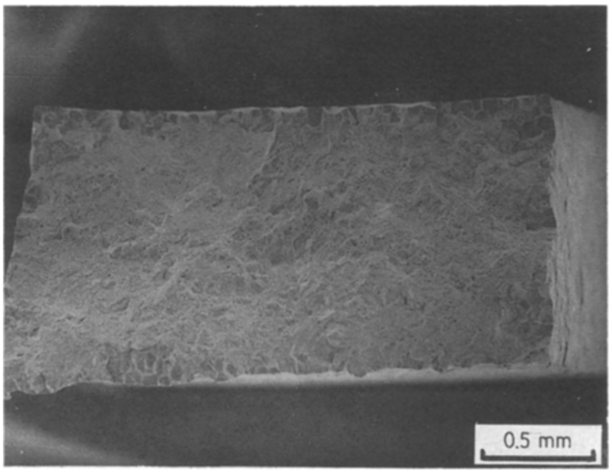

*Figure 10* Fracture patterns of the boron-doped $Ni_3(Si, Ti)$ alloy which was tested at 673 K in air.

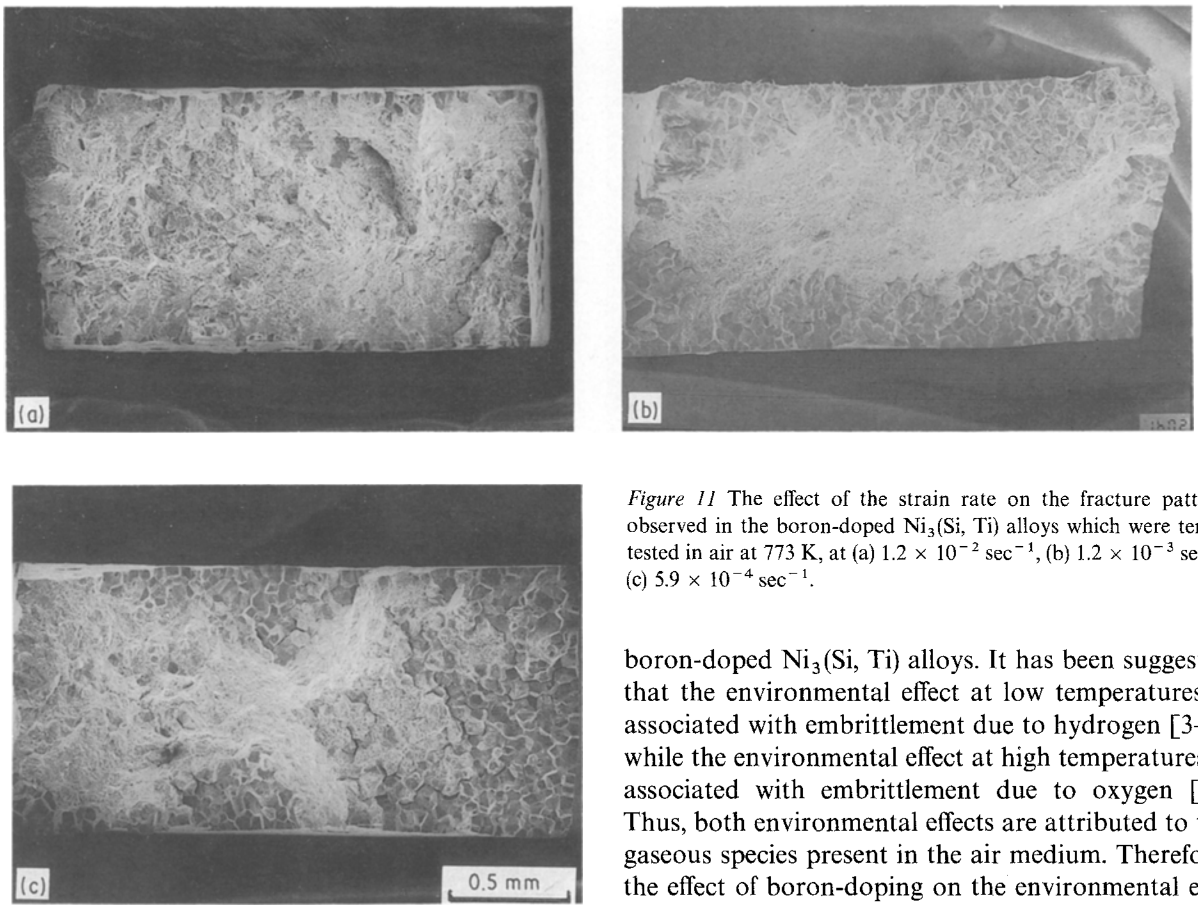

*Figure 11* The effect of the strain rate on the fracture patterns observed in the boron-doped $Ni_3(Si, Ti)$ alloys which were tensile tested in air at 773 K, at (a) $1.2 \times 10^{-2}$ $sec^{-1}$, (b) $1.2 \times 10^{-3}$ $sec^{-1}$, (c) $5.9 \times 10^{-4}$ $sec^{-1}$.

would be influenced by the test temperature and the strain rate. Fig. 10 shows the fracture pattern of this alloy which was tested at 673 K and in air. A comparison of this figure with Fig. 8b clearly indicates that the surface-affected zone (i.e. the intergranular fracture zone affected by the air) introduced at low test temperature is confined to a more limited region. Fig. 11 shows the effect of strain rate on the fracture patterns observed in the boron-doped $Ni_3(Si, Ti)$ alloys which were tensile tested in air and at 773 K. The sample tested at the highest strain rate of $1.2 \times 10^{-2}$ $sec^{-1}$ exhibited a more confined surface-affected zone (Fig. 11a). At the lowest strain rate tested ($5.9 \times 10^{-4}$ $sec^{-1}$), a more extended surface-affected zone was observed (Fig. 11c).

Changes in the fractographies with strain rate were also observed for the boron-doped $Ni_3(Si, Ti)$ alloys which were tensile tested in vacuum and at 773 K. It was shown that in this case the fracture patterns were homogeneous throughout the fractured surfaces of the samples, as shown already in Fig. 8a, and the transgranular fracture patterns were more dominant with increasing strain rate. Similar changes with strain rate were shown in the fractographies of the undoped $Ni_3(Si, Ti)$ alloys which were tensile tested at 573 K and both in vacuum and in air.

## 4. Discussion

Results observed in this study clearly demonstrate that the environmental effect at low temperatures was obvious for the undoped $Ni_3(Si, Ti)$ alloys, whereas the effect was evident at high temperatures for the boron-doped $Ni_3(Si, Ti)$ alloys. It has been suggested that the environmental effect at low temperatures is associated with embrittlement due to hydrogen [3–5] while the environmental effect at high temperatures is associated with embrittlement due to oxygen [6]. Thus, both environmental effects are attributed to the gaseous species present in the air medium. Therefore, the effect of boron-doping on the environmental embrittlements associated with hydrogen or oxygen could be interpreted by an understanding of the interaction between hydrogen and boron or the interaction between oxygen and boron in the corresponding temperature regime.

First, the environmental effect associated with hydrogen which was operative at ambient temperatures will be discussed. The undoped $Ni_3(Si, Ti)$ alloys actually suffered from environmental (i.e. hydrogen) embrittlement. This kind of environmental effect has been widely observed for a number of $L1_2$-type compounds; $Co_3Ti$ [7, 8], $Ni_3(Al, Mn)$ [9] and $Ni_3Al$ doped with beryllium [10]. A micromechanism associated with this phenomenon has been proposed [7, 8]; the dynamic and atomistic mechanism by which the cohesive strength of a grain boundary [7] and the associated plastic flow around a microcrack were affected by hydrogen [8]. As the test temperature increases, the tensile elongation and the UTS of the samples tested in air increased and at 473 K reached the values of samples tested in vacuum. This change with temperature was also investigated in the $Co_3Ti$ alloys which were tested in air [11]. As the test temperature increases, the mobility of hydrogen is supposed to be higher, but the enrichment of hydrogen into a region of a propagating microcrack will be alternatively reduced owing to the entropy effect. As a result, this kind of embrittlement disappears at high temperatures. Naturally, the temperature at which the embrittlement disappears depends on the test strain rate.

On the other hand, the results observed in the boron-doped $Ni_3(Si, Ti)$ alloys clearly indicated that boron atoms prohibited environmental embrittlement in air. Similar beneficial effects of boron have also been investigated in the $Ni_3Al$ alloys; the ductility of the boron-doped $Ni_3Al$ was basically insensitive to the test environment [12, 13] and the test strain rate [13], although this alloy has been embrittled by compulsory injected hydrogen [14]. Therefore, it is again proposed that boron competing with another interstitial atom, hydrogen, for site occupation or for its effectiveness at grain boundaries, has the effect of suppressing the hydrogen embrittlement [4, 13].

The environmental effect associated with oxygen which would be operative at high temperatures was not evident in the undoped $Ni_3(Si, Ti)$. This result is similar to that for $Co_3Ti$ alloys [11]. However, the boron-doped $Ni_3(Si, Ti)$ alloys suffered from this kind of environmental embrittlement. This result is similar to those obtained in the $Ni_3Al$-based alloys doped with boron [6]; the boron-doped $Ni_3Al$ alloy did not represent the environmental effect on the tensile ductilities at room temperature but did represent the environmental effect at high temperatures [6]. At high temperatures ( ~ 600 °C), the boron-doped $Ni_3Al$ polycrystals showed lower tensile ductilities in air than in vacuum. The loss in ductility was accompanied by a change in fracture mode from transgranular to intergranular. It was suggested that this embrittlement was due to a dynamic effect simultaneously involving localized stress concentrations and gaseous oxygen at elevated temperatures. The process of this oxygen embrittlement has been considered to consist of some general steps: (i) adsorption of gaseous molecules, (ii) dissociation of molecules to form atoms or ions, (iii) diffusion and enrichment of oxygen to the stressed region and/or grain boundary (or cracked free surface) near the crack tip, and (iv) loss of mechanical strength.

The environmental effect at high temperatures has thus been observed only in the nickel-based $L1_2$ alloys containing "boron". This fact suggests that the interaction of oxygen and boron is important in this process. One possibility is that the low-melting (eutectic) phase consisting of oxygen and boron (and/or constituent elements of nickel, silicon and titanium) was formed during dynamic loading, resulting in the easier separation of grain boundaries. Indeed, the dendritic phases were observed at grain boundaries near the sample surface. The boron-doped $Ni_3(Si, Ti)$ alloys used in this work contained precipitates of boride at the grain boundaries [2]. Therefore, it is highly likely that the low-melting phase nucleated at these precipitates and then grew, otherwise free boron enriched (segregated) at grain boundaries introduced the new phases. To support this idea, more detailed chemical and structural analysis is required. A second possibility is that the grain boundary cohesion associated with the bonds amongst the constituent atoms is strongly influenced by the co-existence of oxygen with boron.

Finally, a description of the strain-rate effect on the mechanical properties associated with the high-temperature embrittlement is given. As the strain rate decreases, the elongation and the UTS values decreased and correspondingly the portion of the intergranular fracture increased, regardless of the alloy composition and the testing environment. This result reveals that the grain-boundary fracturing at elevated temperature strongly depends on the mobility of the constituent atoms in competition with the applied deformation rate (i.e. the rate of a propagating microcrack along a grain boundary). The diffusivity of atoms at or around grain boundaries to create the grain-boundary cavity or to separate the grain-boundary plane may control the ductility in this temperature regime. It is expected here that as the strain rate decreases sufficient oxygen is provided to the crack tip and/or the crack surface, resulting in more severe intergranular embrittlement.

## 5. Conclusions

The $Ni_3(Si, Ti)$ polycrystals, undoped and boron-doped with 50 p.p.m., were tensile tested over a wide range of test temperatures, and then compared with the results of tensile tests in vacuum [2]. The following results were obtained.

1. The yield stresses were, over all test temperatures, independent of the test environment (vacuum or air).
2. The environmental effect on the undoped $Ni_3(Si, Ti)$ alloys was significant at ambient temperatures but not at elevated temperatures, whereas that for the boron-doped $Ni_3(Si, Ti)$ alloys was not remarkable at ambient temperatures but was so at elevated temperatures. The elongation and the UTS values were reduced at these temperature regimes, although the yield stresses remained constant, when these alloys were tensile tested in air.
3. As the strain rate decreases, the elongation and the UTS values decreased, but the yield stress remained constant, regardless of the doping of boron and the test environment.
4. A correlation was found between fractography and ductility in that as the elongation value (or UTS) increased the fracture pattern changed from the intergranular to the transgranular patterns. When the samples were tensile tested in air and also at low strain rate, the intergranular fracture was dominant.
5. The boron-doped $Ni_3(Si, Ti)$ alloys tested in air and at 773 K showed very heterogeneous fracture patterns; almost intergranular fracture patterns near the sample surface and more transgranular fracture patterns in the sample interior. Also, the dendritic phases were observed at the grain-boundary facets appearing near the sample surface of these alloys after fracturing.
6. It was suggested that the environmental embrittlements at low and high temperatures were due to hydrogen and oxygen absorbed from the air, respectively, and both were caused by a dynamic effect involving the weakening of the grain boundary.
7. At low temperatures, it was proposed that boron competing with hydrogen, for site occupation or for its effectiveness at grain boundaries, has the effect of suppressing hydrogen embrittlement. At high temperatures, however, it was suggested that the low-melting phases consisting of boron and oxygen (and constituent atoms) could be responsible for the loss of ductility in the boron-doped $Ni_3(Si, Ti)$ alloys.

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