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# Interfaces in nickel aluminide/alumina fibre composites

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Metal matrix composites based on the intermetallic alloy $Ni_3Al$ and fibres of $Al_2O_3$ were fabricated by hot-pressing nickel aluminide powders and alumina fibres. Two matrix alloys were used in this investigation: $Ni_3Al$ microalloyed with boron and $Ni_3Al$ alloyed with 8 at% chromium and smaller amounts of zirconium and boron. The materials were studied using optical and transmission electron microscopy with particular emphasis placed on the characteristics of the matrix–fibre interface. The base $Ni_3Al/Al_2O_3$ composite displayed no evidence of chemical reaction at the interface, an intimate bond between matrix and fibre was observed, and the material exhibited 10% ductility at room temperature. Composites with the more complex matrix alloy were brittle, a phenomenon attributed to the formation of zirconia particles at the interface.

## 1. Introduction

Whisker-reinforced metal matrix composites have shown the potential to lead to improved mechanical properties when compared to the unreinforced matrix material. For example, aluminium reinforced with whiskers of silicon carbide shows a dramatic increase in stiffness, yield strength, and ultimate tensile strength [1]. With this in mind, new metal matrix composites based on the intermetallic alloy $Ni_3Al$ and fibres of $Al_2O_3$ have been developed, with the goal to produce materials with favourable high temperature mechanical properties.

Nickel aluminide ($Ni_3Al$) has the unusual property that its yield strength increases with increasing temperature. When $Ni_3Al$ with substoichiometric aluminum ($\sim 24$ at %) is microalloyed with boron, it exhibits room temperature ductilities of up to 50% [2]. Although $Ni_3Al$ embrittles in an oxygen atmosphere at high temperature, the addition of over 8 at % chromium has been shown to alleviate this problem [3]. Thus, a boron-doped $Ni_3Al$ with additions of chromium appears to be an excellent candidate for the matrix material in a high temperature composite. Because prior investigations have shown that nickel aluminides chemically react with silicon carbide to form nickel silicides, alumina fibres were chosen as the reinforcement [4].

In this investigation, the effects of the $Al_2O_3$ fibres on the matrix microstructure were examined by optical microscopy and transmission electron microscopy (TEM). In addition, the matrix–fibre interface was characterized by TEM, with particular attention directed toward finding evidence of chemical reaction at the interface. Preliminary tensile and bend tests were carried out at room temperature and the fracture modes were characterized.

## 2. Experimental

Two matrix alloys with composition Ni–24.0 Al–0.24 B (at %) (Oak Ridge National Laboratory (ORNL) designation IC-15) and Ni–16.5 Al–8.0 Cr–0.4 Zr–0.1 B (ORNL designation IC-218) were used in this investigation. The zirconium was added to IC-218 to improve the creep resistance. Polycrystalline alumina fibres ($\alpha$-$Al_2O_3$) 20 $\mu$m in diameter and 3 mm in length were obtained from the E.I. DuPont de Nemours Co. (DuPont designation FP Type I). Composites were fabricated by mixing the nickel aluminide powders (nominally 100 $\mu$m diameter) with 20 vol % alumina fibres in a hexane slurry with a high shear mixer. The mixtures were dried and then hot-pressed in graphite dies. To obtain close to theoretical density, a temperature of 1300–1350° C using 20 MPa pressure for 1 h was needed for the IC-15 composite, and a temperature of 1250–1300° C using 20 MPa pressure for 1.5 h was sufficient for IC-218/$Al_2O_3$ material. Samples of each composite were then annealed for 2 h at 1000° C and 24 h at 800° C to relieve residual stresses in the material. Additionally, powders of the IC-15 and the IC-218 alloys were hot-pressed without fibre reinforcement under the same conditions as their respective composites so that alloy microstructures could be compared.

Specimens for optical metallography were prepared by mechanical polishing and light etching. Specimens of the composite materials were prepared for TEM by mechanically polishing a 3 mm diameter disc to approximately 25 $\mu$m thickness and then ion milling

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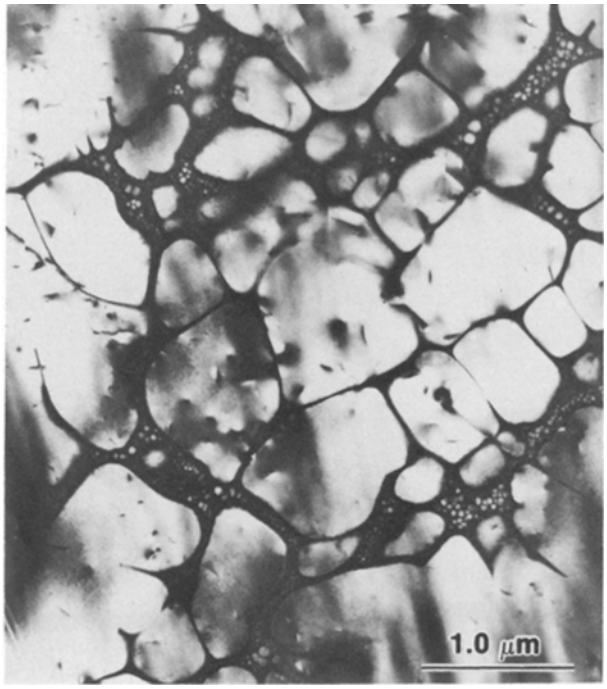


*Figure 1* Superlattice dark field transmission electron micrograph of hot pressed IC-218 (Ni–16Al–8Cr–0.4Zr–0.1B) powder showing disordered web (dark regions) running through the ordered matrix. Ordered precipitates can be seen inside the disordered web.

to perforation. Specimens of the hot-pressed alloys were electropolished at $-12°C$ using a solution of 58.5 vol% methanol, 35% 2-butoxy ethanol, 6% perchloric acid, 0.5% glycerin as the electrolyte. TEM studies were done using a Philips EM400T equipped with a field emission gun or with a Philips EM430T and an EDAX 9100/70 energy dispersive X-ray spectrometer (EDS).

Four-point bend tests were performed on both composite materials and the powders pressed without fibres to provide a simple and reliable estimate of ductility. The test setup could accommodate strains of up to 5%. Tensile tests were sometimes used to verify the ductility. Tests were performed in air at room temperature on the IC-15/$Al_2O_3$ composite. The specimens tested had a gage section of 12.7 × 3.18 × 0.76 mm [3].

## 3. Results and discussion

The following materials were examined by optical and transmission electron microscopy: both base alloys in the as-cast condition, both composites after hot-pressing with the alumina fibres, and the nickel aluminide powders after hot-pressing but without the alumina fibres. Microstructural observations of the cast material, the powder product, and the composite after hot-pressing were compared with particular regard to the formation of any precipitates.

The base nickel aluminide (IC-15) after casting and homogenization was single phase, completely ordered with the $L1_2$ structure with no precipitates present. The modified nickel aluminide alloy (IC-218 with chromium and zirconium added) had a more complex microstructure, characterized by ordered regions separated by a web of disordered material similar to that shown in Fig. 1. Similar structures in nickel aluminides with iron additions have been described previously [5]. No precipitation involving zirconium was observed indicating that the zirconium was still in solution.

TABLE I Compositions of hot-pressed IC-218 powder (at %)

| | Ni | Al | Cr | Zr |
|---|---|---|---|---|
| Ordered matrix | 76.0 | 16.1 | 7.9 | < 1 |
| Disordered | 77.1 | 5.3 | 17.6 | < 1 |
| Nominal (IC-218) | 74.0 | 16.0 | 8.0 | 0.4 |

The rapid solidification process associated with powder production results in the production of an antiphase boundary structure with an associated change in aluminium-to-nickel ratios from grain centre to grain boundaries [6]. When the aluminium level equals or exceeds 25%, a martensitic $\beta$ phase forms [7]. Rapid solidification of Ni–Al–Cr alloys also has been reported but the aluminium plus chromium level was much higher than in the present investigation [8]. In the alloys used here, the aluminium or aluminium plus chromium levels were kept below 25%.

The two unreinforced powder metallurgy, P/M, alloys examined by TEM showed substantially different microstructures. The hot-pressed IC-15 powders without fibre reinforcement had an average grain size of 50 $\mu$m and contained a low dislocation density. However, the hot-pressed IC-218 powders exhibited two phases and had a grain size of 10 $\mu$m. The phase separation was attributed to the addition of chromium [9], and the observed microstructure resembled that of the cast alloy. Dark field images ($\bar{g} = 110$) of the hot-pressed IC-218 powder revealed ordered regions which appeared bright and disordered regions that were dark (Fig. 1). A web of disordered material surrounded regions of ordered material within the grains, and no misorientations were detected across the webs. Within the disordered web a reprecipitation process has occurred during the high temperature hot-pressing. These small ordered precipitates within the disordered web were not present in the cast alloy after an anneal at 1000° C for 2 h followed by 24 h at 800° C. Compositions of the phases were determined by EDS and are shown in Table I. The analysis of IC-218 showed that the disordered regions were chromium enriched and aluminium depleted, while the composition of the ordered phase was closer to the nominal composition. In addition, small zirconia ($ZrO_2$) particles (identified by electron diffraction), 150–300 nm in diameter, were often observed at grain boundaries in the hot-pressed IC-218 alloy. The formation of zirconia particles resulted from the reaction of zirconium in solution with excess oxygen that was present during hot-pressing.

TEM observations of the fibres showed fine equiaxed grains ($\sim 0.5$ $\mu$m diameter), and cavities present in the alumina. The cavities were uniformly dispersed throughout the fibres, suggesting that they were present before hot-pressing the composites. The surface of the fibres was microscopically rough because of the random shapes of the alumina grains.

The optical micrographs in Fig. 2 show the effect of the alumina fibres on the microstructure of IC-15 and

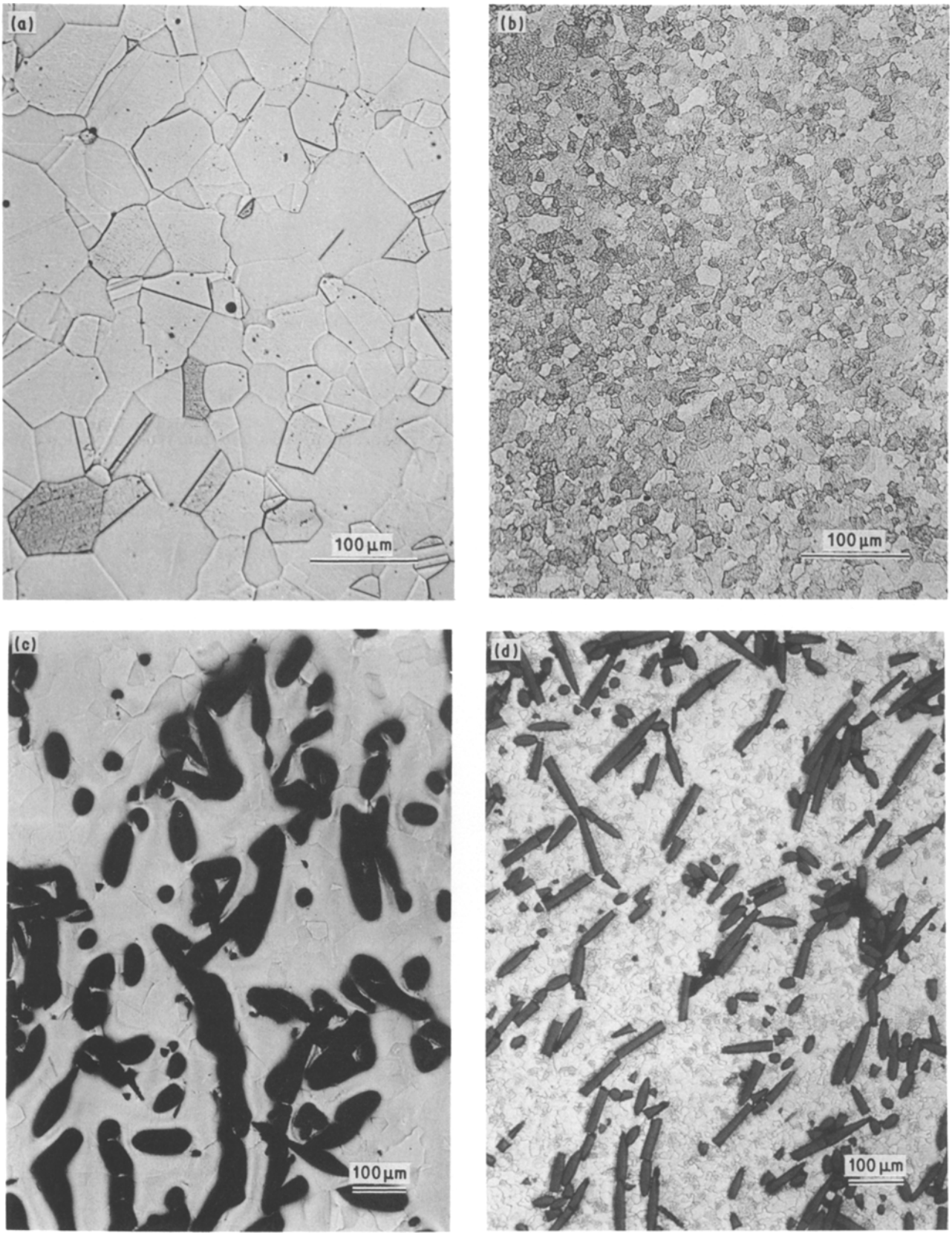


*Figure 2* Optical microstructures of hot-pressed nickel aluminides and $Ni_3Al/Al_2O_3$ composites. (a) IC-15, Ni–24Al–0.24B, grain size 50 $\mu$m. (b) IC-218, Ni–16Al–8Cr–0.4Zr–0.1B, grain size 10 $\mu$m. (c) IC-15/$Al_2O_3$ composite, matrix grain size 25 $\mu$m. (d) IC-218/$Al_2O_3$ composite, matrix grain size 7 $\mu$m.

IC-218 matrix alloys. The presence of the fibres tended to inhibit grain growth especially in the IC-15 material, resulting in smaller grain sizes in the matrices of the composites. Although some fibre clusters were observed, in general the fibres were uniformly dispersed and randomly oriented. Substantial fibre breakage occurred during the processing of the composite, resulting in a reduction of the mean aspect ratio of the fibres from the original 150 : 1 to between 7 : 1 and 15 : 1.

The matrix in both composite materials had high dislocation densities, even after the material had been annealed. Dislocations were particularly dense near the matrix–fibre interface, but were still prevalent in

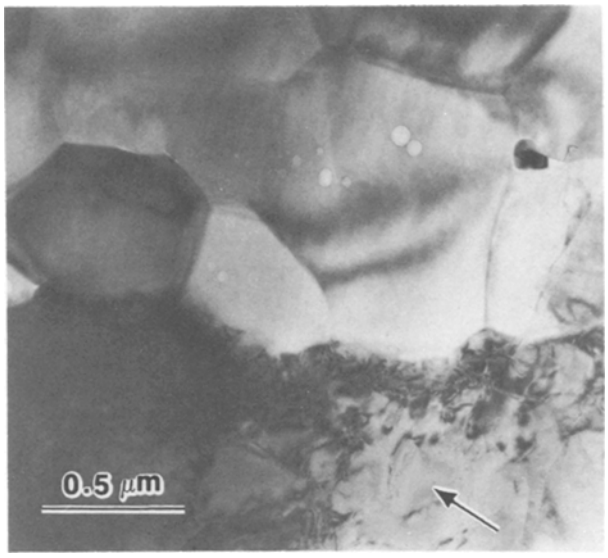


*Figure 3* Bright field transmission electron micrograph of IC-15/$Al_2O_3$ interface. Arrow points to the IC-15 matrix.

grains with no fibres in the vicinity. However, matrix alloys hot-pressed without fibres had much lower dislocation densities. The different thermal expansion coefficients of the matrix and the fibres ($12.5 \times 10^{-6}$ and $6.5 \times 10^{-6}\,°C^{-1}$ at room temperature and $16.8 \times 10^{-6}$ and $12.0 \times 10^{-6}\,°C^{-1}$ at 1000° C respectively) causes large thermal stresses to develop upon cooling. The thermal stresses apparently exceed the matrix yield stress, thereby causing plastic flow, a phenomenon that has also been observed in fibre-reinforced aluminum composites [10, 11]. This presumed effect is also complicated by the increased difficulty in annealing out dislocations due to the increased pinning at the interfaces. The major implication of this observation is that the matrix is in a work-hardened state, and the mechanical properties of the composites should be influenced accordingly. No evidence of porosity in either matrix material was observed.

Figs 3 and 4 show matrix fibre interfaces for both the IC-15 and IC-218 composites. In both materials, no continuous reaction phases between the nickel aluminide matrix and the alumina fibres were observed

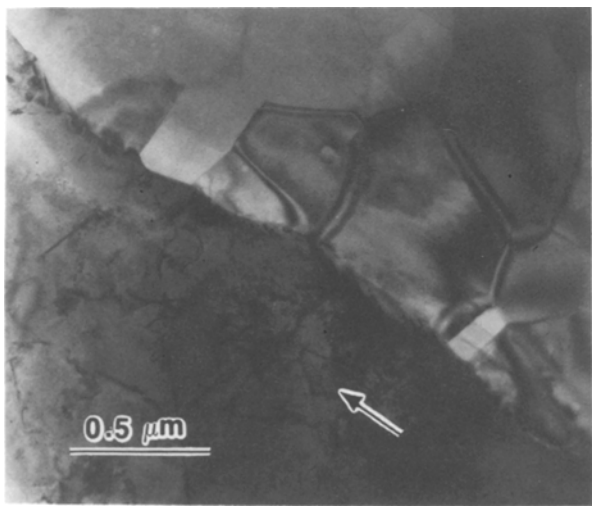


*Figure 4* Bright field transmission electron micrograph of IC-218/$Al_2O_3$ interface. Arrow points to the IC-218 matrix.

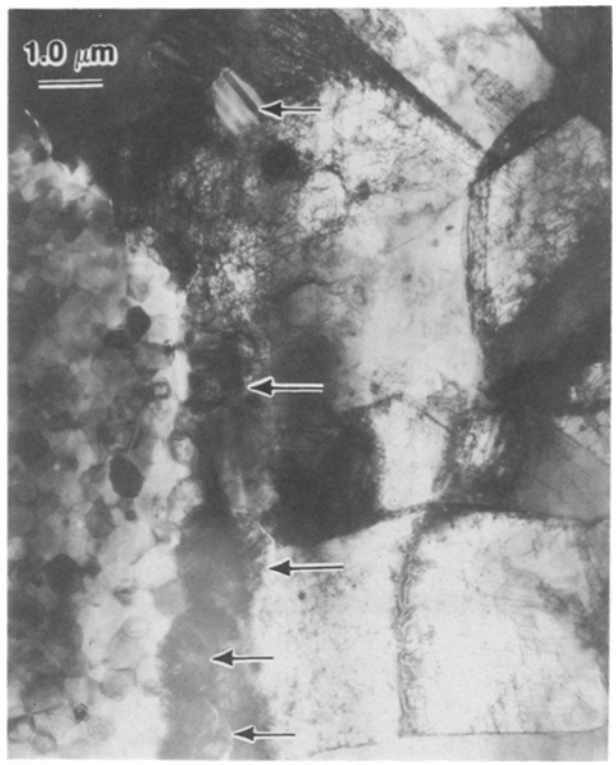


*Figure 5* Bright field transmission electron micrograph of an annealed IC-218/$Al_2O_3$ composite showing zirconia particles on an alumina fibre. Cracking between the particles and the matrix is evident. Arrows point to the zirconia particles.

along the interface. Concentration profiles measured across the interfaces by EDS using a probe size of 5 nm revealed no evidence of interface segregation in either composite. Similarly, microdiffraction patterns showed no evidence of any reaction between the alumina fibres and the nickel aluminide alloys. The cohesive strength of the interface could not be determined from TEM, but the fact that the matrix was in intimate contact with the fibre surface and had not separated during specimen thinning suggests that bonding had occurred.

Zirconia particles observed in the unreinforced IC-218 powders were also present in the IC-218 composites, although the distribution was different. Zirconia particles 1–2 μm in diameter were present along the matrix–fibre interface (Fig. 5). The unusually large size of the $ZrO_2$ particles (10 times the size of the particles in the unreinforced IC-218 alloy) was attributed to enhanced nucleation at the fibre surface. Cracks were often observed between the $ZrO_2$ particles at the interface and the IC-218 matrix, indicating the bond between the particles and the fibres was stronger than the bond between the particles and the matrix. In contrast, composites with the IC-15 alloy matrix which contained no zirconium showed no signs of cracks. Zirconia particles in the IC-218 matrix away from the interface were the same size as those in the unreinforced IC-218 alloy. Note that the smaller zirconia particles were not detrimental to the material without fibres while the presence of larger particles in the reinforced material is associated with increased cracking and lower ductility. Compositions of the two types of zirconia particles as determined by EDS


*Figure 6* Scanning electron micrograph showing a fracture surface of an IC-15/$Al_2O_3$ composite.

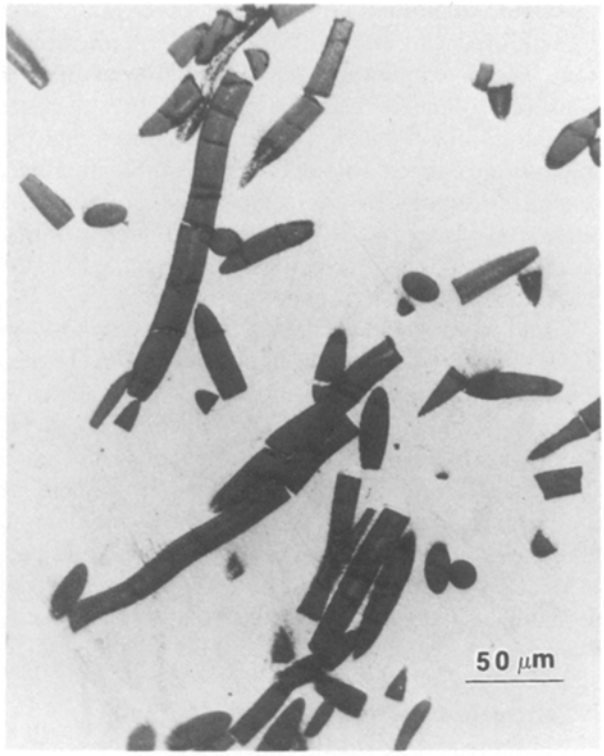


*Figure 7* Optical micrograph of IC-15/$Al_2O_3$ composite strained 10% in air at room temperature. Note the cracks in the fibres.

analysis was similar. The addition of alumina fibres did not, however, alter the distribution of ordered and disordered material in the matrix.

Annealed specimens of the IC-15/$Al_2O_3$ composite material were tested in tension at room temperature in air. The material showed an elongation to failure of 10%. Examination of the fracture surface revealed mixed modes of failure, including fibre breakage, interface decohesion, and transgranular cracking through the matrix (Fig. 6). Most of the fibres oriented transverse to the tensile axis debonded from the matrix, while those fibres parallel to the tensile axis tended to fracture, and fibres inclined to the tensile axis showed evidence of both mechanisms. Metallographic examination of sections taken from the composite fracture specimens showed that most fibres had developed transverse cracks and were bent (Fig. 7). Cracks were not observed in sections of as-pressed composites, and fibres were generally straight (Fig. 2), indicating that the cracks were not induced by mechanical polishing. Closer inspection by TEM revealed that secondary microcracks were initiated in the matrix in the vicinity of a fibre crack (Fig. 8). The fibre crack did not propagate directly across the interface into the matrix, and many small matrix cracks were initiated from the interface nearby. The matrix cracks were typically about 1 $\mu$m and extended normal to the fibre interface. Fibre cracks generated during the tensile test were apparently deflected along the interface without propagating across the matrix.

Four-point bend tests performed on the IC-15/$Al_2O_3$ composites resulted in measured yield strengths of up to 550 MPa, an increase of up to 48% compared to the unreinforced P/M alloy [12]. The ductilities of both were greater than 5% (the limit of the bend test specimen holding apparatus). Consequently tensile tests at room temperature were used to verify the ductility. These tests of IC-15/$Al_2O_3$ yielded ductilities of 10%. Bend tests conducted on the IC-218/$Al_2O_3$ material resulted in brittle failure with a maximum strain of less than 1%. The observed low ductility was attributed to the pre-existence of cracks between the large zirconia particles at the interface and the metal matrix (Fig. 5). The fracture stress was approximately 170–200 MPa. In comparison, the IC-218 P/M alloy exhibited yield stresses in bending of up to 900 MPa with ductilities of greater than 5% [12].

## 4. Summary

Composites fabricated by hot-pressing powders of the base nickel aluminide alloy (IC-15) with alumina

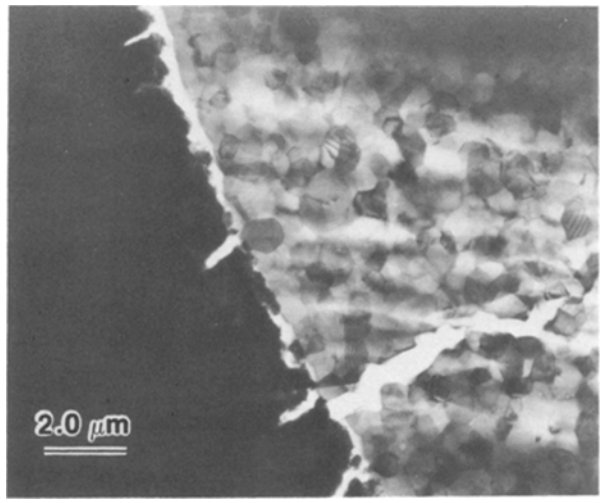


*Figure 8* Bright field transmission electron micrograph of an IC-15/$Al_2O_3$ composite strained 10% in air at room temperature showing small cracks in the matrix initiated by a fractured alumina fibre.

fibres showed intimate contact between the matrix and the reinforcement and no evidence of interface reaction. Room temperature ductility of the composite measured by uniaxial tensile tests was 10%. Tensile deformation of composite specimens showed that the cracks often deflected along the matrix–fibre interface, generating matrix microcracks in the process. The observation suggests that at room temperature, a nickel aluminide matrix is sufficiently ductile to resist cracks introduced from fibre breakage.

Composites using a matrix alloy of composition NI–16.5 Al–8.0 Cr–0.4 Zr–0.1 B (IC-218) were also studied. Chromium is present because of its effect of improving the high temperature ductility of nickel aluminides in an oxidizing environment, while small additions of zirconium increase the creep resistance of the alloy. Hot-pressing of these powders with alumina fibres resulted in the formation of large zirconia particles at or near the fibre interface. These particles did not bond well with the $Ni_3Al$ matrix, and consequently the material exhibited poor ductility.

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