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## THE EFFECT OF MICROSTRUCTURE ON DUCTILITY OF NiAl

### WPŁYW MIKROSTRUKTURY NA PLASTYCZNOŚĆ NiAl

NiAl, an intermetallic compound with the B2 ordered structure, exhibits potential as a viable high temperature structural material. However, before this material (and other intermetallics) can be of practical use a number of technical problems must be overcome, including lack of ductility at the room temperature. In an attempt to address the brittleness of NiAl, and based on a premise that grain refinement may lead to significant ductility improvements mechanical alloying and nanocrystalline synthesis were used to produce fine-grained NiAl. The mechanically alloyed NiAl with an average grain size of about 0.5  $\mu\text{m}$ , unlike its coarse grained, cast counterpart, exhibits a notable room temperature compressive ductility due to two contributing microstructural factors: i) the development of the  $\langle 110 \rangle$  texture during hot extrusion leading to the activation of additional slip systems, and ii) the predominance of low angle grain boundaries. In the nanocrystalline form, NiAl with an average grain size in the range from 2 to 10 nm, exhibits measurable room temperature ductility in biaxial disc bend tests, unlike its coarse-grained counterpart. This observation can be explained assuming that diffusional, rather than dislocation, mechanisms control plastic deformation of the nanocrystalline NiAl. The emphasis of the present paper is on rationalizing the improved room temperature ductility in mechanically alloyed and nanocrystalline NiAl. The most significant conclusion of the present discussion – contrary to widespread beliefs – is that the grain size plays only an indirect role in controlling ductility.

NiAl, związek międzymetaliczny o strukturze krystalicznej B2 jest potencjalnym materiałem do zastosowań konstrukcyjnych w wysokich temperaturach. Jednakże, zanim ten materiał (a także inne intermetaliki) znajdzie praktyczne zastosowanie muszą zostać pokonane liczne techniczne przeszkody, włączając w to kruchość w temperaturze pokojowej. Opierając się na założeniu, że rozdrobnienie ziarna może prowadzić do znacznej poprawy plastyczności NiAl, wytworzono ten związek w drobnoziarnistej postaci metodami mechanicznej oraz nanokryst-

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talicznej syntezy. Mechanicznie wytwarzany NiAl, o wielkości ziarna ok. 0,5  $\mu\text{m}$ , wykazywał znaczną plastyczność podczas próby ściskania w temperaturze pokojowej, w przeciwieństwie do NiAl wytwarzanego klasyczną metodą wytapiania. Przyczynami wzrostu plastyczności w materiale wytwarzanym mechanicznie były: i) rozwój tekstury włóknistej  $\langle 110 \rangle$  podczas wyciskania „na gorąco”, która ułatwia aktywację dodatkowych systemów poślizgu, oraz ii) duży udział w strukturze granic niskokątowych. W nanokrystalicznej formie o wielkości ziarna ok. 2 do 10 nm, NiAl wykazywał w temperaturze pokojowej mierzalną ciągliwość w dwuosiowej próbie zginania, natomiast materiał o większym ziarnie był kruchy. Obserwacja ta prowadzi do wniosku, że dyfuzyjne, a nie dyslokacyjne mechanizmy kontrolują odkształcenie plastyczne NiAl w nanokrystalicznej formie. W niniejszej pracy położono nacisk na wyjaśnienie zjawiska poprawy plastyczności NiAl w temperaturze pokojowej wywołanego rozdrobnieniem ziarna. Najważniejszym osiągnięciem tej pracy jest wykazanie, wbrew ogólnemu przekonaniu, że zmniejszenie wielkości ziarna w NiAl wpływa jedynie w pośredni sposób na plastyczność.

Keywords: intermetallics, mechanical alloying, nanocrystalline materials, ductility, grain size.

## 1. Introduction

Intermetallic compounds, such as nickel and titanium aluminides, are under active consideration as a new class of potential structural materials for high temperature applications such as gas turbine engines. Among others, NiAl, an intermetallic compound with the B2 ordered bcc crystal structure, exhibits potential as a high temperature structural material, either in a monolithic form or as a matrix phase in a composite, because of its low density, high melting temperature, good thermal conductivity and excellent oxidation resistance [1]. However, before this material (and other intermetallics as well) can be of practical use a number of technical problems must be overcome, including, first and foremost, very limited or even lack of ductility at the room temperature. Several attempts to resolve the problem of the room temperature brittleness through modification of slip systems [2], grain refinement [3], grain boundary elimination, and microalloying with boron [4], have been made, however, with limited successes. The brittleness of intermetallic materials remains one of the greatest challenges in the field of high temperature structural materials [5].

In the present paper we focus on: i) discussing the potential of using grain refinement to improve room temperature ductility of NiAl, and ii) interpreting and rationalizing apparent progress that has been made in the last ten years or so by an Illinois Institute of Technology (IIT) research group led by one of the co-authors (M. Dollár). The two processing techniques used were: i) mechanical alloying (leading to grain refinement and slip system modifications [6], and ii) processing of nanostructured materials (resulting in extremely fine grains in the nanometer range [7]).

## 2. Mechanically alloyed NiAl

### 2.1. Processing and characterization of mechanically alloyed NiAl

Mechanical alloying followed by hot extrusion was used to produce fine grained NiAl-based alloys (containing oxide dispersoids) in an attempt to address the room temperature brittleness (and, in fact, high temperature strength problems). Mechanically alloyed (MA) NiAl was obtained from elemental Ni and Al powders in a Szegvari attritor mill. MA powders, collected after milling, were sieved to  $45\ \mu\text{m}$  (-325 mesh), degassed at 973K for 1.5 hours in a vacuum furnace, encapsulated under vacuum in a stainless steel can and hot extruded at 1400K at a ratio of 16:1. A near-stoichiometric NiAl was prepared for comparison with the MA material from a cast ingot and hot extruded under the same conditions.

Optical and transmission electron microscopy (TEM) studies of the hot-extruded MA NiAl revealed fine, equiaxed grains with an average size of about  $0.5\ \mu\text{m}$  and a fairly homogenous distribution of alumina oxide dispersoids throughout the matrix (Fig. 1).

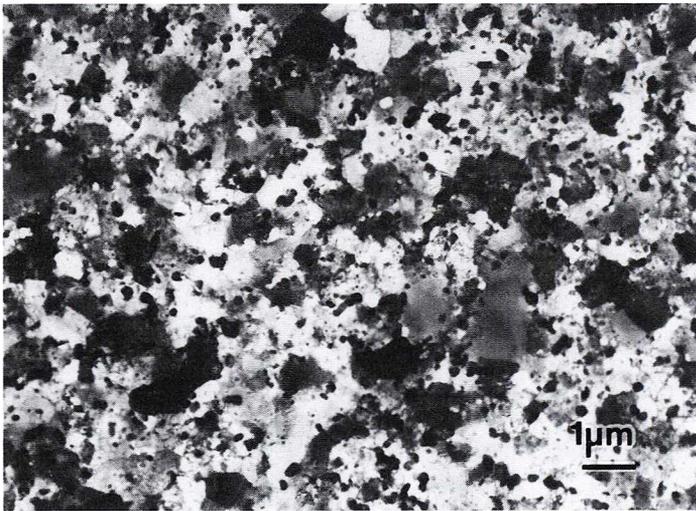


Fig. 1. Transmission electron micrograph of the hot-extruded MA NiAl

The optical and transmission electron microscopy observations showed that the cast NiAl is a single phase material with an average grain size of about  $30\ \mu\text{m}$  (Fig. 2).

The  $\{110\}$  and  $\{200\}$  pole figures were analyzed to give orientation distribution functions and inverse pole figures. The analysis indicates that the MA NiAl exhibits a strong  $\langle 110 \rangle$  fiber texture parallel to the extrusion axis while the hot extruded cast material has a  $\langle 111 \rangle$  fiber texture (Figs. 3 and 4). The  $\langle 110 \rangle$  texture is a common

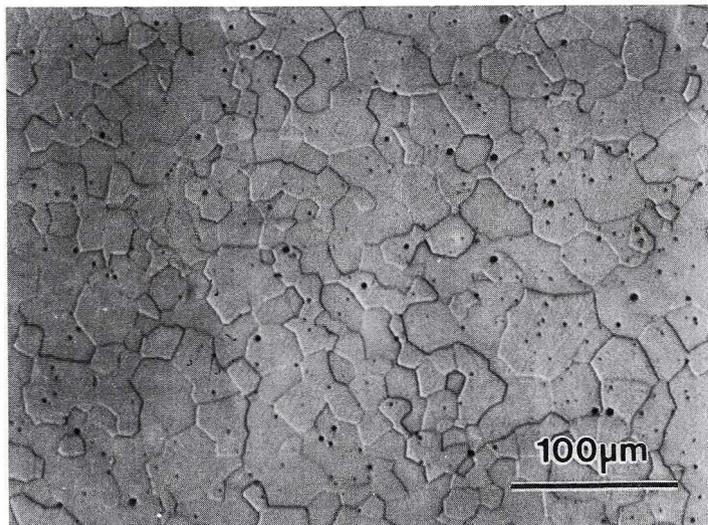


Fig. 2. Optical micrograph of the hot-extruded cast NiAl

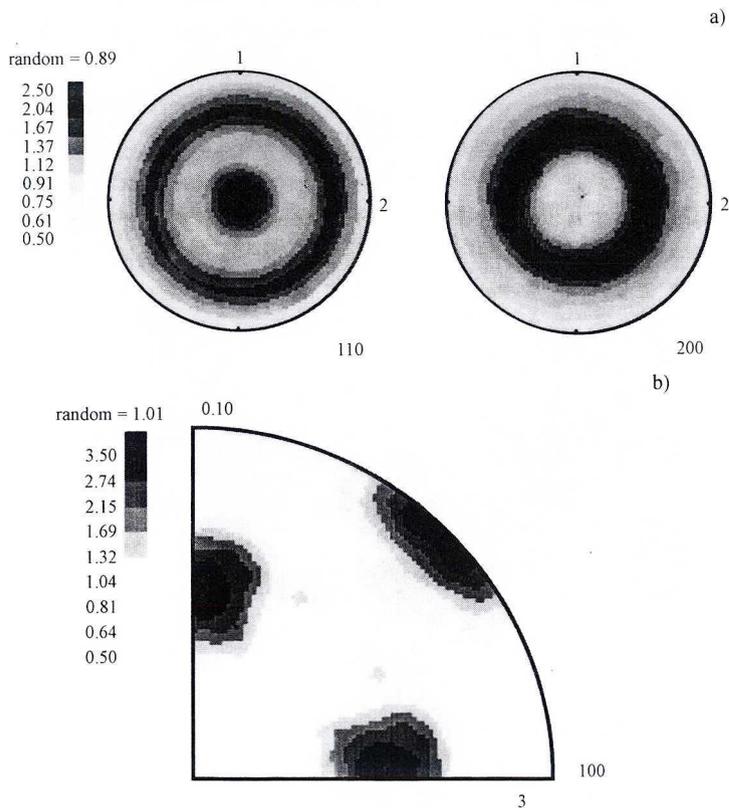


Fig. 3. (110) and (200) pole figures (a) and the corresponding inverse pole figure (b) of the hot-extruded MA alloy

texture produced by extrusion (or cold drawing) in body-centered cubic materials. Since the texture of MA powders is essentially random and featureless, the  $\langle 110 \rangle$  texture in MA NiAl is a direct consequence of the deformation during extrusion. This deformation texture is postulated to be retained in MA NiAl alloys due to the presence of dispersoids. The mean distance between the oxides is much less than a critical recrystallization nucleus size (typically about  $1 \mu\text{m}$ ) and the nucleation is prevented altogether. The cast material, in turn, was subjected to stress and high temperature simultaneously during hot extrusion, and dynamic recrystallization took place. In fact, the microstructure of the as-extruded cast NiAl consists of recrystallized and equiaxed grains. A common recrystallization texture observed in the cast NiAl is  $\langle 111 \rangle$  [8], the observation confirmed in the study in question.

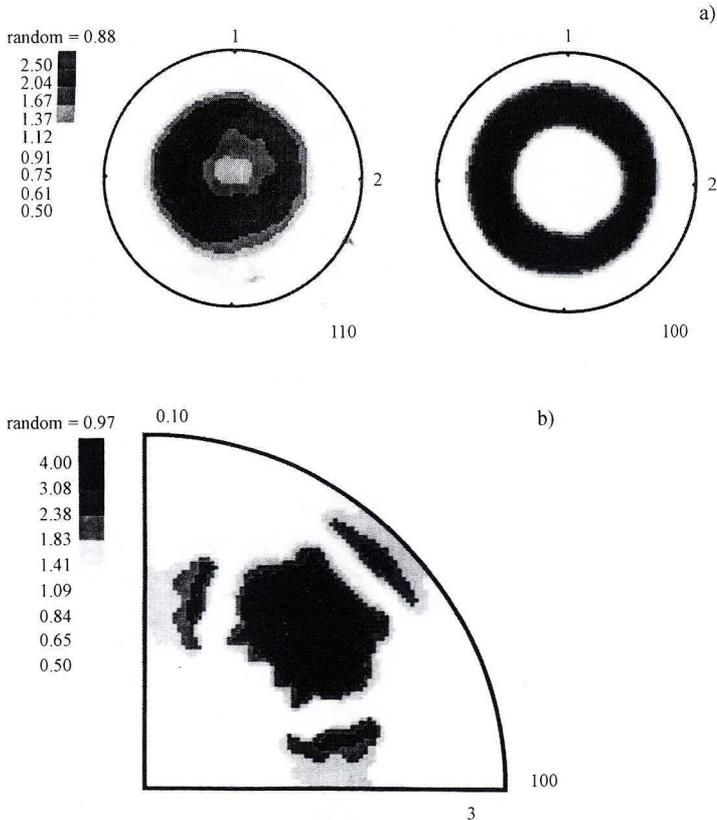


Fig. 4. (110) and (200) pole figures (a) and the corresponding inverse pole figure (b) of the hot-extruded cast alloy

## 2.2. Ductility of MA NiAl

Several specimens of cast and mechanically alloyed NiAl were subjected to compression tests. The strain till failure of the cast NiAl was in all the specimens below 3%. The compression tests of MA materials had to be stopped when the load on the specimen reached the limit of the Instron cell. When the machine was stopped, the compressive strains in the mechanically alloyed specimens were typically about 12% at the room temperature.

NiAl is an aluminide with the ordered B2 cubic structure. Several studies have been undertaken to determine the operative slip systems at the room temperature in NiAl. The consensus of these investigations is that this compound deforms predominantly by  $\{110\} \langle 100 \rangle$  slip [9]. The occurrence of  $\langle 100 \rangle$  slip was confirmed in the present MA and cast NiAl.

The  $\{110\} \langle 100 \rangle$  slip, typically observed in NiAl compound at the room temperature, provides only three independent slip systems, as first predicted by Copley [10]. No additional slip systems are provided by cross-slip, since the three operative slip directions are mutually at right angles [11]. The occurrence of only three slip systems is believed to be responsible for the room temperature brittleness of NiAl, as the number of independent slip systems available for general deformation is insufficient in the light of the generally accepted von Mises criterion for plasticity [12] requiring at least five independent slip systems in a grain for general plasticity. The role of slip systems availability is reinforced when one realizes that polycrystalline NiAl intermetallics exhibit a brittle to ductile transition in the temperature range 300 to 600°C, depending on composition, grain size and processing [14]. This transition is believed to be the result of the activation of new slip systems with vectors  $\langle 110 \rangle$  and  $\langle 111 \rangle$  at elevated temperature [14,15]. In the research studies in question, only  $\langle 100 \rangle$  slip vectors were observed in the cast NiAl at room temperature, and, not surprisingly, this material exhibited very limited compressive ductility.

In the MA NiAl, the occurrence of  $\{110\} \langle 001 \rangle$  slip was confirmed and, more importantly, strong evidence for the  $\langle 110 \rangle$  slip direction was obtained [16]. The occurrence of the  $\langle 110 \rangle$  slip in the MA alloy is believed to be a result of relatively strong  $\langle 110 \rangle$  fiber texture which enables the activation of  $\{110\} \langle 100 \rangle$  and  $\{110\} \langle 110 \rangle$  slip systems while the  $\langle 111 \rangle$  fibre texture in the cast NiAl allows only  $\{110\} \langle 100 \rangle$  slip systems to operate. To determine the number of independent slip systems in the present situation the method, proposed by Groves and Kelly [17] was followed [16]. It was proven that when the  $\langle 001 \rangle$  and  $\langle 011 \rangle$  slip vectors are encountered, five independent slip systems operate and the von Mises requirement for general plasticity is satisfied. The operation of five independent slip systems is suggested to contribute to the notable compressive ductility of the MA NiAl-based materials.

Another factor likely contributing to the improved compressive ductility in the MA NiAl is the predominance of low angle grain boundaries (the factor also associated with the strong texture), facilitating the transmission of slip between neighboring grains (Fig. 5), and likely not allowing large incompatibility stresses to develop in their vicinity.

The differences in compressive ductility between cast and MA NiAl could also be attempted to be attributed directly to the difference in grain size. Grain refinement was in fact suggested to be a way of obtaining ductility in NiAl [3]. Limited evidence indicates that grain size effects on ductility can be quite significant. For example, it was reported that while the ductility of NiAl at 400°C was very low (about 2% tensile elongation) and independent of grain size  $d > 20 \mu\text{m}$ , at finer size the ductility increased with decreasing grain size [4]. NiAl thus appears, at least at 400°C, to exhibit a critical grain size below which it becomes

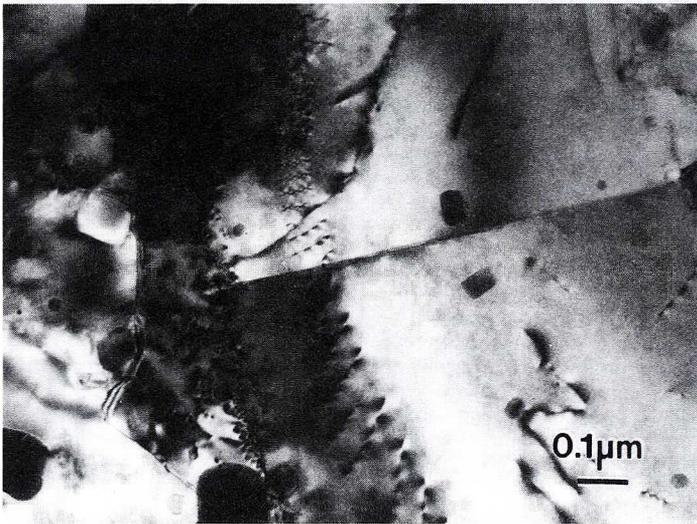


Fig. 5. Transmission of slip across a low angle grain boundary in the MA alloy; 2% deformation by compression at room temperature

ductile in tension. Cottrell's concept [18] that a critical grain size exists below which the stress to nucleate cracks is less than the stress to propagate them may help to explain this behavior. In fine-grained polycrystals, in terms of the Cottrell's notion, crack propagation proceeds only after plastic flow has occurred. However, the adoption of this concept in the present case would oversimplify the problem since it does not account for the different deformation mechanisms in fine-grained MA NiAl.

Marginal ductility, of the order of a few tenth of 1%, was observed in MA NiAl in tension, suggesting that processing flaws causing premature tensile failure were present.

### 3. Nanocrystalline NiAl

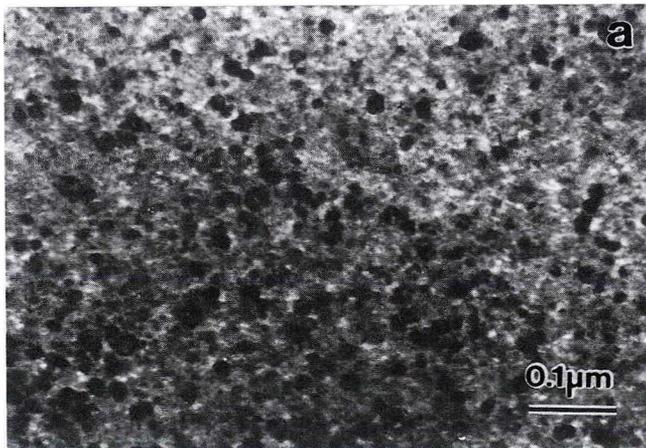
#### 3.1. Processing and characterization of nanocrystalline NiAl

As described in detail elsewhere [7], the nanocrystalline NiAl (n-NiAl) specimens in the form of 9 mm diameter discs were synthesized by inert gas condensation, using an electron beam physical vapor deposition system. This efficient method allows to synthesize powders of high melting point, multi-component materials. In this study, nickel and NiAl were evaporated from separate crucibles with electron beam dwell times chosen to yield the desired 5% Ni – 50% Al composition. A nickel source was required to compensate for a large vapor pressure of Al compared to Ni. The powders were compacted at 1.4 GPa, at temperatures ranging from 100 to 300°C, into 9 mm-diameter disk-shaped specimens. The specimens exhibited densities ranging from 78% to 94% of the theoretical density, and the density increased with increasing compaction temperature. The specimens with the highest density were chosen for further discussion.

Transmission Electron Microscopy (TEM) bright and dark field images along with a corresponding diffraction pattern, representative of the n-NiAl, are shown in Fig. 6 a–c. The observed grains are randomly oriented, essentially equiaxed, with sizes between 2 and 10 nm. The rings are observed in the diffraction pattern despite the fact that the small selected area aperture (400 nm) was used; the aperture though is still large enough to encircle hundreds of nanometer-size grains.

*In-situ* TEM studies proved that the nanostructure of n-NiAl is stable up to 1000°C. There is very little grain growth up to 400°C and a modest increase between 400 and 1000°C. The content of Ni was found to be between 46 and 53 at.% well within the range of the occurrence of the ordered phase. The Vickers microhardness of the as-compacted n-NiAl ranged from 5000 to 5500 MPA whereas that of the coarse-grained NiAl is typically in the range from 2800 to 3100 MPA.

The detailed discussion of the synthesis and consolidation processes and a full characterization of the as-compacted specimens are presented elsewhere [19].



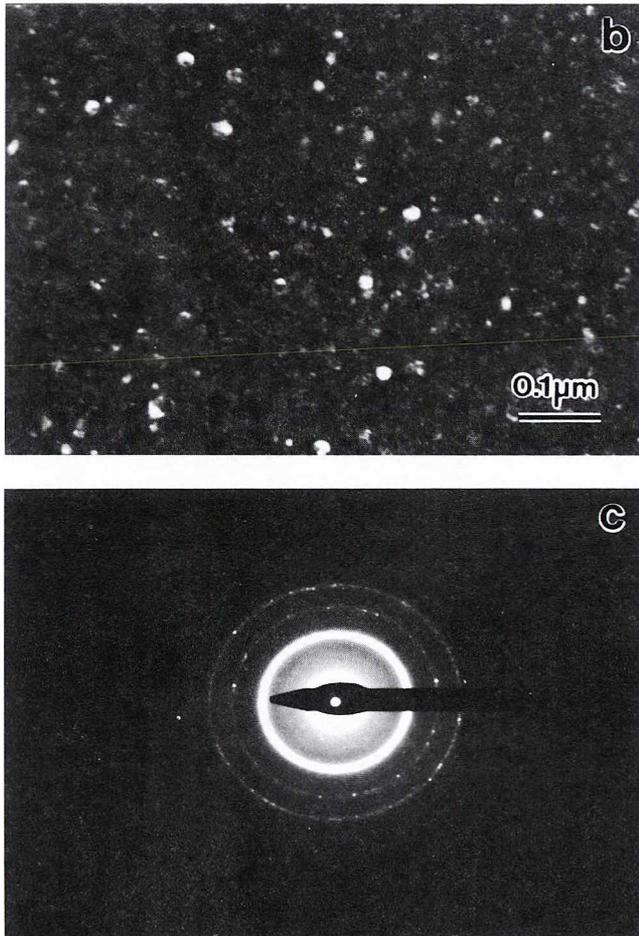


Fig 6. Transmission electron microscopy images of the nanocrystalline NiAl a) bright field image, b) dark field image, c) selected area diffraction pattern

### 3.2. Ductility of n-NiAl

Biaxial Disc Bend (BDB) tests were conducted using the as-compacted n-NiAl specimens to investigate the room temperature behavior of the material. Coarse-grained cast NiAl specimens of the same geometry were used as reference material. The BDB tests allowed to test as-compacted specimens and eliminated the need for specimen machining. System calibration and performance evaluation were done by testing standard stainless steel and aluminum specimens. Yield stresses and elastic moduli measured were within  $\pm 6\%$  of the literature values for the same materials obtained in conventional tensile tests. Figures 7 and 8 show representative load – displacement

curves for a coarse-grain NiAl specimen and a n-NiAl specimen respectively subjected to the BDB test. No plastic yielding was observed in the coarse-grained specimens. A notable deviation from linearity was observed in the curves for n-NiAl providing an evidence of room temperature ductility in NiAl in the nanocrystalline form: Strains-to-failure of 1.0 to 0.3% were determined. SEM studies of postmortem specimens revealed a ductile failure mode n-NiAl as opposed to a brittle mode in the conventional NiAl.

It is well known that metallic materials in the nanocrystalline range are significantly less ductile than their coarse-grained counterparts [20]. The brittleness of nanocrystalline metals (n-metals) can be rationalized assuming that dislocation generation is increasingly difficult as the grain size decrease. In fact La salmonie and Strudel estimated the limitations of grain-size strengthening models based on dislocation slip; for the pile-up model the lower limit was found to be about 100 nm whereas for a dislocation source model *ca.* 15 nm [21]. This is believed to lead to a loss of dislocation-controlled ductility in n-metals. On the other hand, diffusional mechanisms are expected to be more active in n-metals given their higher volume fractions of grain boundaries, as well as triple junctions [22]. Estimations, reviewed by Hauboldt *et al.* [23], of the basic equations for diffusion-controlled mechanisms such as Nabarro-Herring, Coble, Ashby-Verall creep and grain boundary sliding mechanisms indicate that grain boundary diffusion leading to grain boundary sliding is expected to be the mechanism controlling low temperature deformation in n-metals. However, in n-metals the increase in ductility provided by diffusional mechanisms is apparently insignificant compared to the loss of dislocation controlled ductility. In contrast to the behavior of n-metals, the experimental evidence reviewed in this paper indicates that in the case of a material

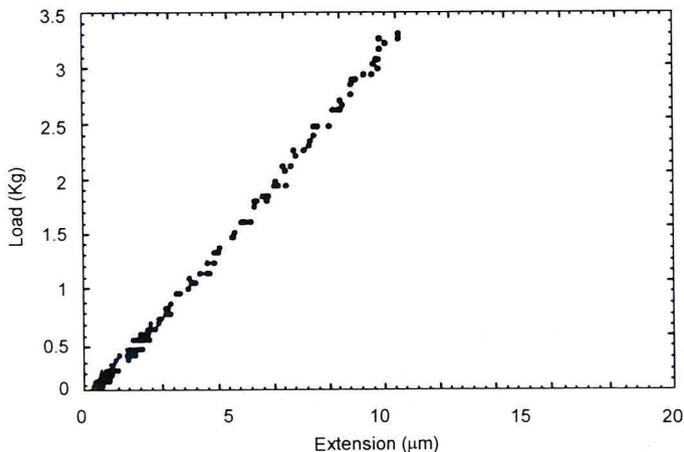


Fig. 7. Load displacement plot of coarse-grained NiAl tested at room temperature

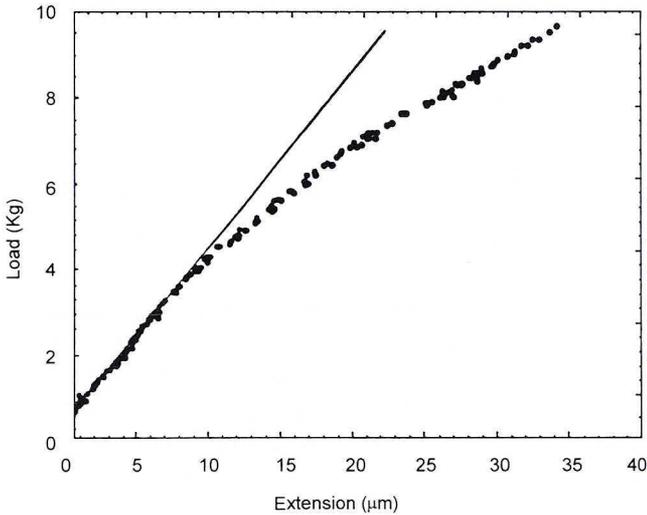


Fig. 8. Load displacement plot of nanocrystalline NiAl tested at room temperature

brittle in coarse-grained form such as NiAl, enhancement of diffusional mechanisms accompanying grain refinement results in a measurable room temperature ductility. This notion is reinforced by the fact that in a material with the grain size of the 5 to 10 nm range approximately 20% of the materials volume is occupied by grain boundaries [20].

#### 4. Summary

Whereas conventionally processed, cast NiAl has grains with an average size of about 30  $\mu\text{m}$ , the two processing methods in question mechanical alloying and nanocrystalline synthesis, result in significant grain refinement. The grain sizes produced by those processing routes are in the range of a few tenths of a micron and a few nanometers, respectively. The grain refinement leads to the improved ductility. The cast NiAl is brittle at the room temperature in both compression and tension while the MA NiAl shows the significant compressive ductility. The n-NiAl, in turn, unlike its cast counterpart, exhibits a measurable ductility in the BDB tests.

The improved ductility, though clearly not sufficient if NiAl is to be used in load bearing applications, seems to reinforce the notion that the ductility of otherwise brittle materials can be directly controlled by grain refinement. The notion, for-

mulated in the late 80's, was based on the above-mentioned classic Cottrell's concept [18] that a critical grain size exists below which the stress to nucleate cracks is less than the stress to propagate them, and as a result, crack propagation proceeds only after plastic flow has occurred. However, in light of the present discussion, it is clear that the grain size plays only an indirect role in controlling ductility.

In the MA NiAl, the dispersoids affect the progress of recrystallization and contribute to the preservation of the  $\langle 110 \rangle$  deformation texture. The  $\langle 110 \rangle$  texture enables the activation of  $\langle 100 \rangle\{110\}$  and  $\langle 110 \rangle\{110\}$  slip systems. The occurrence of  $\langle 100 \rangle$  and  $\langle 110 \rangle$  slip dislocations satisfies the von Mises criterion of general plasticity and is postulated to contribute to the notable compressive ductility of the MA materials. Another factor likely affecting the compressive ductility is the predominant occurrence of low angle grain boundaries facilitating the transmission of slip between neighboring grains. In the n-NiAl, enhancement of diffusional mechanisms in the material in which approximately 20% of the materials volume is occupied by grain boundaries results in a measurable room temperature ductility.

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