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MECHANICAL PROPERTIES OF NANOCRYSTALLINE METALS, INTERMETALLICS AND MULTIPHASE MATERIALS DETERMINED BY TENSION, COMPRESSION, AND DISK-BEND TECHNIQUES*

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Abstract

The mechanical behavior of nanocrystalline metallic, intermetallic, and multiphase materials has been investigated using tension, compression, and disk-bend techniques. Nanocrystalline NiAl, Al-Al_3Zr, and Cu were synthesized by the gas condensation technique using either resistive or electron beam heating followed by elevated temperature vacuum compaction. Disk-bend tests of nanocrystalline NiAl show evidence of improved ductility at room temperature in this normally extremely brittle material. In contrast, tension tests of multiphase nanocrystalline Al-Al_3Zr samples show significant increases in strength but substantial reductions in ductility with decreasing grain size. Compression tests of nanocrystalline copper result in values of yield stress and total elongation that are substantially larger than those measured in tensile tests. Implications for the operable deformation mechanisms in these materials are discussed.

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Introduction

During recent years numerous efforts have been made to characterize the mechanical behavior of a variety of nanocrystalline materials. For example, it is well established that nanocrystalline metals exhibit much greater microhardness values than their coarse-grained counterparts [1-6]. Tensile testing of normally ductile metals such as Pd and Cu [6-10] has shown that large increases in yield strength are obtained with decreasing grain size, but that in general a significant loss of ductility accompanies this strengthening. Flaws that can arise during the synthesis of nanocrystalline samples have been shown to have a deleterious effect on the strain-to-failure of nanocrystalline metals [6-10]. Even after improving synthesis techniques and thus reducing the prevalence of flaws [11], very limited ductility is still observed [9, 10]. Whether this indicates that flaws are still controlling the strain-to-failure or that nanocrystalline metals are intrinsically limited in ductility is not yet clear.

Most previous studies have used primarily microhardness measurements and/or tensile testing to characterize the mechanical behavior of nanocrystalline metals. A smaller number of studies have utilized other testing techniques such as compression or disk-bend testing. Compression testing may be useful in characterizing more accurately intrinsic grain-size dependent mechanical properties since this technique is expected to be less susceptible to the influence of flaws than tensile testing. However, compression testing has only been employed in one previous study that we are aware of [12], perhaps because much larger samples are typically required for compression testing than for tensile testing. Suryanarayanan *et al.* [12] reported that the yield strength measured in compression of nanocrystalline Cu with a mean grain size of approximately 40 nm and a density of about 90% of theoretical is approximately double that of similarly grain-sized Cu investigated in tension by Nieman *et al.* [6].

Disk-bend testing [13, 14] also offers possible advantages over tensile testing in characterizing nanocrystalline samples, particularly since the thin disk-shaped samples typically produced by the gas condensation synthesis process can be tested in as-produced form. This method eliminates the need to machine dogbone-shaped samples and subsequently attach grips or strain gauges to the tiny dogbones. A disadvantage of disk-bend testing, however, is that the data analysis is less straight-forward than for uniaxial tension or compression tests due to the more complex stress states. Disk-bend testing was used by Gertsman *et al.* [15] to characterize the yield strength of severely deformed Cu with grain sizes ≥ 170 nm and it was observed that, consistent with tensile test results, significant increases in yield strength accompany reduction in grain size.

The goals of the studies described in this report were threefold: 1) to extend previous tensile testing studies to investigate multicomponent alloys, in this case Al-Al_3Zr [16], 2) to more accurately determine the intrinsic mechanical properties of clean, high density nanocrystalline Cu by performing compression tests on thick samples [17], and 3) to use disk-bend testing to determine whether a normally brittle material such as NiAl can be made more ductile by reducing grain size to the nanocrystalline scale [18]. Taken together, the results of these three studies allow one to compare and contrast the deformation mechanisms operating in nanocrystalline materials that are typically either ductile (Cu and Al-Al_3Zr) or brittle (NiAl) in coarse-grained form. By utilizing tensile, compression, and disk-bend techniques in these studies, the roles of flaws and stress states on mechanical behavior are also probed.

Results

Tensile Properties of Nanocrystalline Aluminum-Zirconium Alloys

Nanocrystalline aluminum-zirconium samples with average zirconium concentrations varying from 0 to 6.4 wt.% were produced using the inert-gas condensation technique with electron-beam evaporation [19], followed by uniaxial compression (1.4 GPa) at 100°C and under high vacuum conditions [11]. The densities of samples used for tensile tests were above 96% of theoretical. The synthesis process is described in greater detail elsewhere [16, 20]. Uniaxial tensile tests of polished nanocrystalline dogbone specimens were conducted at a strain rate of 10^{-4} sec^{-1} in displacement control. A description of the sample preparation, tensile testing, and strain measurement procedures is given in [21].

As reported previously [22], the microstructures of the Al-Zr samples consist of nanocrystalline aluminum, the majority phase, and nanometer-sized particles of other phases, primarily the cubic Al_3Zr structure. Shown in Figure 1 are engineering stress-strain curves obtained from tensile tests of three nanocrystalline samples and coarse-grained aluminum sheet (99.998% purity). The TEM dark field micrographs adjacent to the curves show the grain size distribution in each of the nanocrystalline specimens tested. Each nanocrystalline sample was consolidated under identical conditions at 100°C, and the difference in resulting grain sizes can be linked to the different levels of zirconium in each sample. Sample A contained on average 6.4 ± 0.3 wt.% Zr; sample B, 0.6 ± 0.3 wt.% Zr; and sample C contained no zirconium. Sample A had a measured average grain size of approximately 9 nm. Sample B exhibited a bimodal grain size distribution, with some regions of the sample having also approximately a 9 nm grain size and other regions having grain sizes of a few hundred nm. Sample C had a uniform average grain size of a few hundred nm, as seen in Fig. 1.

The stress-strain data in Fig. 1 show a correlation between ductility level and grain size; the ductility of the samples dropped drastically as the grain size was reduced into the nanometer regime. In turn, the grain size is correlated with zirconium concentration since zirconium additions stabilize the microstructure against grain growth. The percentage of total elongation of the coarse-grained aluminum sample was >50%, and this value dropped to ~12% in a pure aluminum sample with a grain size on the order of a few hundred nanometers. In the two nanocrystalline samples tested containing increasing amounts of zirconium (0.6 and 6.4 wt.%) and correspondingly decreasing grain sizes, the elongation values were only ~2.5 and 0.3%, respectively. In addition to observing a loss of ductility at higher zirconium levels and smaller grain sizes, a significant increase in yield strength was observed. Reduction of the aluminum grain size to a few hundred nanometers resulted in a four-fold increase in the 0.2% offset yield strength (to 120 MPa) compared to that of coarse-grained aluminum (30 MPa). Zirconium additions, which served to further reduce the grain size of the samples, led to

Table 1. Compilation of tensile test results on nanocrystalline Al-Zr samples.

Sample	Ave. Zr content (wt.%)	Modulus (GPa)	Yield Strength (MPa)	Ultimate tensile strength (MPa)	Strain to failure (%)
A	6.4	74	>250	>250	0.3
B	0.6	63	180	243	2.5
C	0	69	120	138	12
coarse-grained Al	0	70	30	41	50

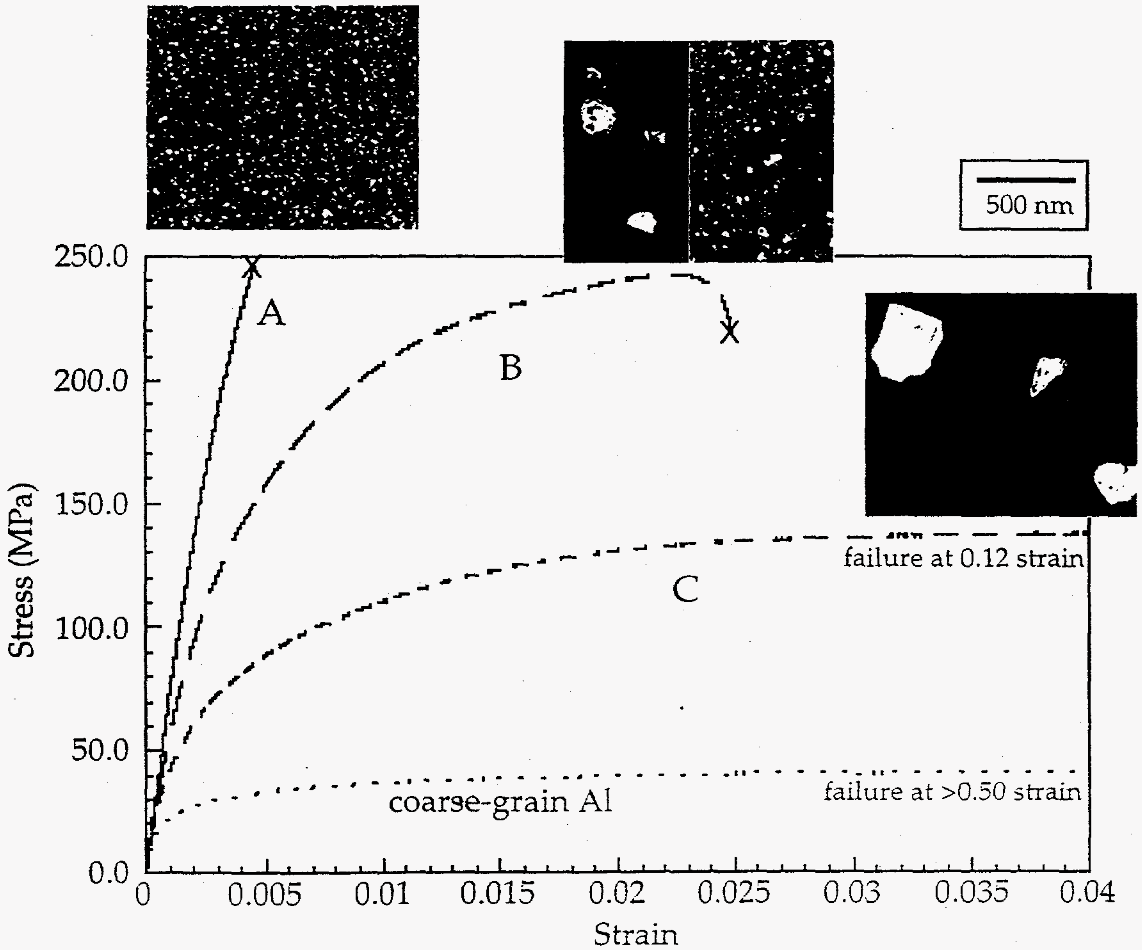

Figure 1. Stress-strain curves obtained from tensile tests of coarse-grained Al and nanocrystalline samples with Zr concentrations ranging from 0-6.4 wt.%. Dark-field electron micrographs for each sample are shown next to the associated tensile data. Note that sample B had a bimodal grain size distribution and that the average grain sizes of the different samples were A < B < C < the coarse-grained sample.

significantly higher yield stresses: 180 MPa in the case of sample B (0.6 wt.% Zr), and >250 MPa in the case of sample A (6.4 wt.% Zr). Because sample A exhibited brittle failure prior to yielding, 250 MPa is actually a lower bound for the yield strength of this nanocrystalline specimen. The mechanical properties obtained from the tensile experiments are compiled in Table 1.

Compression Studies of Nanocrystalline Copper

Mechanical evaluation of nanocrystalline materials, particularly of yield strength, is often achieved via tension testing [6-10]. Due to the small amount of material produced by the commonly used gas condensation synthesis technique (typically less than 1 gram per sample for laboratory systems), 9 mm diameter disk-shaped nanocrystalline samples tend to be thin (< 1 mm in thickness). Unfortunately, the nature of tension testing is such that as sample size decreases, difficulties due to flaws or handling of samples increase. Potential stress concentrators on the sample surface such as the edges of a dogbone sample must be removed on an increasingly fine scale. On many occasions, such finishing work or even the simple process of loading the sample into the testing apparatus results in breakage of the sample.

One way to circumvent this difficulty is to test samples in compression, where flaw size is unimportant. During compression testing, inclusions are loaded and cracks are closed. Compression testing also allows for a simpler sample shape (a cylinder rather than a dogbone). Also, compression testing is less susceptible to the effects of flaws on concentrating local stresses to higher than the macroscopically measured levels, leading to premature yielding or fracture in tension tests. There are, however, two major problems inherent to compression testing. One is friction between the compressing platens and the sample. Friction prevents the top and bottom regions of the sample from deforming linearly with the rest of the sample, resulting in barreling of samples. A certain degree of this effect is normal and acceptable; the way to insure a consistent and negligible effect is to use specified aspect ratios for the sample. A height-to-diameter ratio of 1.5:1 is considered ideal. A second major obstacle to valid compression testing is attaining a precise alignment of the apparatus. Though a tensile sample and jig are self-aligned during the initial stages of a tension test, a misaligned compression apparatus will tend to become more misaligned as the sample is pressed between the platens.

In the past, compression testing was not an option, because samples produced by gas condensation were typically too thin to attain the desired height-to-diameter ratio with samples large enough to handle. While a flat dogbone specimen could be cut from a typical disk (0.1-1 mm thick, 9 mm in diameter), there was no way to produce a valid compression sample over 1 mm in any dimension, a configuration that would prove most difficult to test. While lack of sufficient powder was in some cases a limitation in producing sufficiently thick samples for compression testing, a typically more serious problem resulted from difficulties in transferring all the powder produced successfully into the die for compaction while maintaining vacuum conditions. Recent improvements in the processing method have allowed for the production of samples up to 4.5 mm in thickness [17, 23]. From these samples, 3 mm disks can be cut and a valid configuration obtained. By using a special alignment fixture and a careful finishing procedure, complications with alignment and friction were eliminated. For the current compression tests, nanocrystalline copper powder was synthesized by gas condensation in a resistively heated system, scraped, and delivered to a compaction unit. There the copper powders were compressed under 10 tons (1.4 GPa) of load at 100 °C and at high vacuum (≤ 5E-6 Torr).

A typical engineering stress-strain curve for a compression test of nanocrystalline copper is shown in Figure 2. The grain size of this sample was 19 nm, and it was > 92 % dense. While a similar copper sample may exhibit a tensile strength from 200-400 MPa [6-10], all specimens cut from this sample had 0.2% offset yield stresses of at least 650 MPa. Also, these samples showed greater than 2% ductility, which is significantly larger than the up to 1% ductility observed for samples produced in the same synthesis chamber and tested in tension [9, 10]. Compression tests on other samples give similar or better results, and hardness values are similar to those of samples tested in tension for a given grain size [17, 23]. These results suggest that compression testing overcomes some of the obstacles of tension testing and yields measurements more intrinsic to the material.

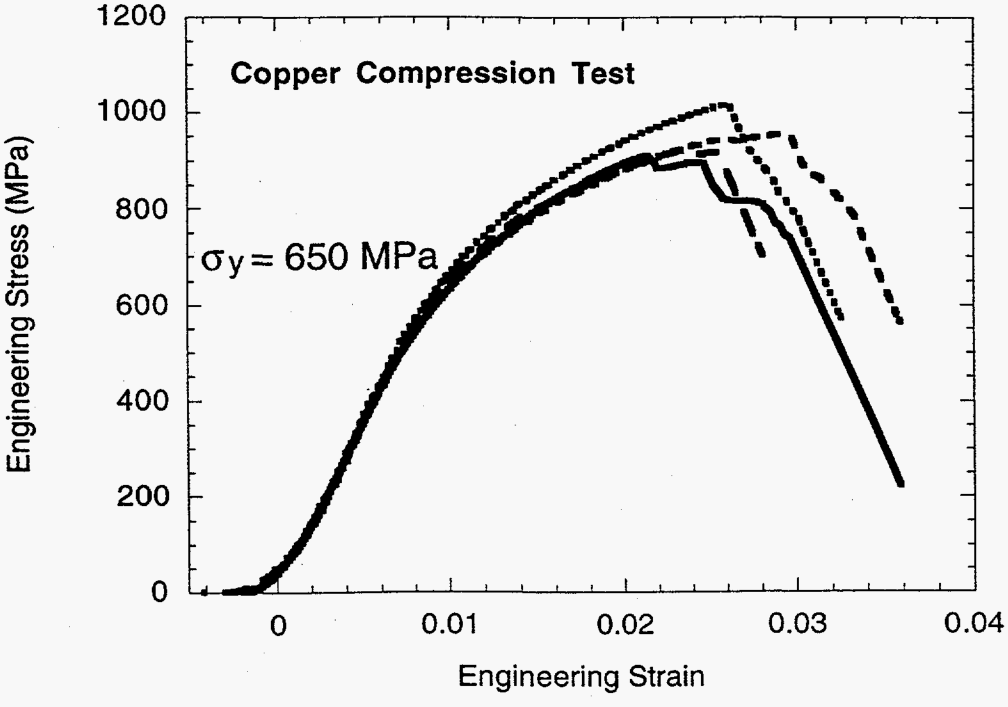

Figure 2. Engineering stress-strain curves for nanocrystalline Cu samples tested in compression at room temperature. The samples had an average grain size of 19 nm and were approximately 92% of bulk density. All four samples were cut from the same original 4.5 mm thick disk and the similarity of the curves indicates the reproducibility of the compression testing technique. All samples exhibited 2-3% strain prior to failure.

Disk-bend Tests of Nanocrystalline NiAl

Intermetallic alloys are of interest for a variety of potential applications because of their high strength-to-weight and stiffness-to-weight ratios and because they often possess excellent elevated temperature properties. The fundamental limitation in the commercial utilization of these materials invariably is their inherent ambient temperature brittleness, which adversely affects material handling and fabricability. A number of recent studies have begun investigating the possibility of improving room ductility through grain size refinement to the nanocrystalline regime. For example, both Chang *et al.* [24] and Xiao *et al.* [25] have produced nanocrystalline TiAl. Chang *et al.* [24] used microhardness measurements to investigate the mechanical behavior of nanocrystalline TiAl and concluded that some evidence of ductilization was observed in their material.

In the present investigation [18], nanocrystalline NiAl samples with average grain sizes below 10 nm were produced using an electron beam evaporation system [19]. High vacuum compactions at temperatures of 100 - 300°C were performed, resulting in sample densities that varied from 70 - 90% of theoretical. Biaxial disk-bend tests were performed to characterize the mechanical behavior of both nanocrystalline and coarse-grained NiAl samples. All samples were mechanically polished to produce a 0.05 μm surface finish. A compression cage fixture was designed and built for these measurements and data were analyzed following the

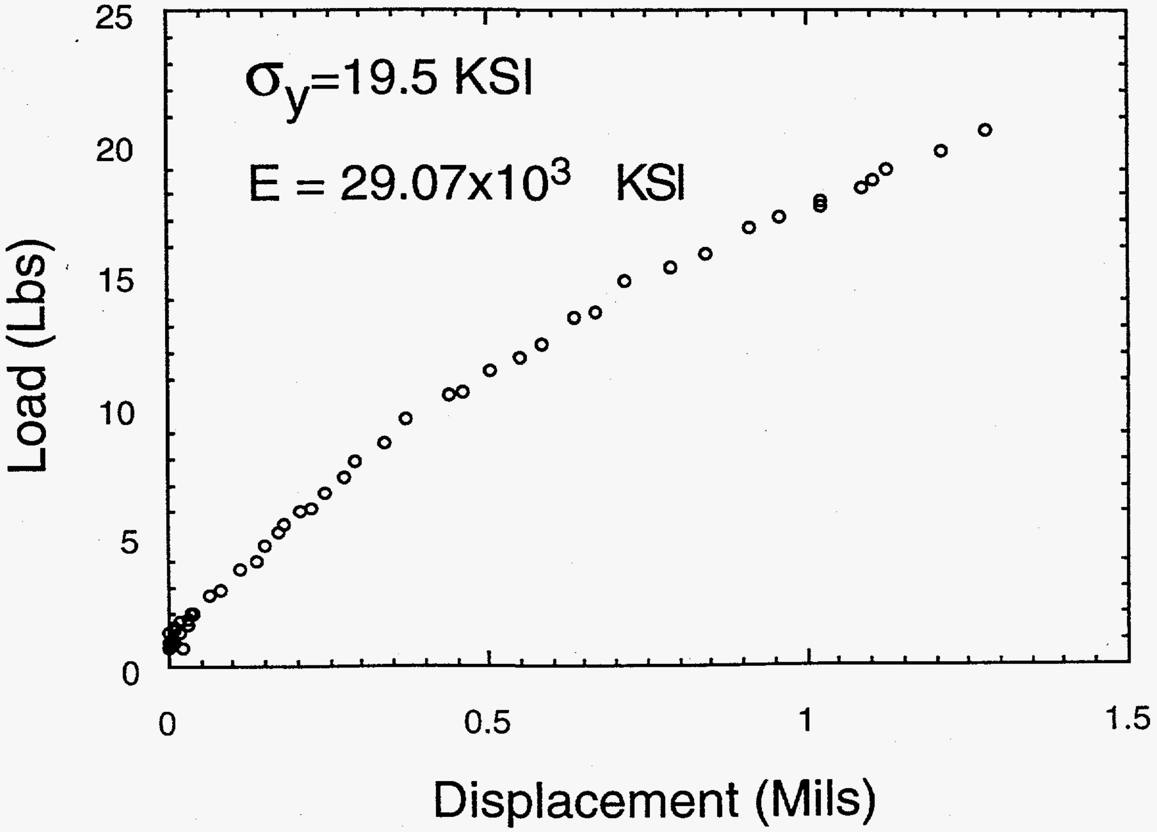

Figure 3. Applied load versus displacement for a nanocrystalline NiAl sample tested by disk-bending at ambient temperature. The deviation from linearity of the data indicates that the sample exhibits measurable ductility.

procedures described in [13]. To ensure the reliability of the measurement technique, coarse-grained stainless steel and Al samples were tested since these materials have well-characterized mechanical properties.

A representative load-displacement curve for a nanocrystalline NiAl sample is shown in Figure 3. After correction for the expected initial non-linear region [13], the value of the total load at yielding was used to calculate the yield stress according to the procedures outlined in [13]. It has been shown by Ardell and co-workers [13] that for disk-bend tests plastic yielding occurs at the onset of the deviation from linearity in the load-displacement curve and that the yield strength obtained by disk-bend testing is comparable to the tensile yield strength. A significant observation in the present study is that a deviation from linearity is observed in the load-displacement curve of Figure 3, indicating that nanocrystalline NiAl exhibits significant room temperature ductility. A deviation from linearity was not observed when coarse grained NiAl was tested under similar conditions.

Discussion and Conclusions

An explanation for the significant increase in strength and corresponding loss of ductility with decreasing grain size in nanocrystalline Cu and Al-Zr alloys comes from examining the operable deformation mechanisms in these materials. Coarse-grained fcc metals are, of course, very ductile, and deformation occurs by dislocation generation and motion. As has been pointed out by numerous authors (e.g., [6]), dislocation generation is increasingly difficult as grain size decreases, leading to a loss of dislocation-based ductility in nanocrystalline materials. Likewise, diffusional mechanisms are expected to be more active in nanocrystalline materials given their higher fractions of grain boundaries, as well as triple- and higher-order junctions [26], all of which display far higher diffusion rates than the bulk. It has been suggested, for example by DiMelfi [27], that grain boundary sliding and diffusional creep mechanisms may be enhanced in nanocrystalline materials under the proper combinations of grain size, test temperature, and strain rate. It is apparent in the cases of Cu and Al-Al_3Zr, however, that the increase in ductility afforded by diffusional mechanisms is insignificant compared to the competing loss of ductility due to the hindrance of dislocation motion under the experimental conditions used. It is possible that more significant ductility could occur under other combinations of test temperature and strain rate, but a detailed exploration of these parameters was beyond the scope of this study.

In contrast to the behavior of materials that are ductile in coarse-grained form, the present studies indicate that in the case of a normally brittle material, such as NiAl, deformation of nanocrystalline samples is likely affected to a measurable extent by enhancements of diffusional mechanisms that accompany grain size refinements. Also, it is improbable that dislocation-based deformation would be enhanced by decreasing the grain size for any material. One aspect of this improved ductility that has not yet been probed in detail is the effect of test method (i.e., disk-bend testing versus tensile or compression testing) on stress-strain curves. Since the stress state during a disk-bend test is complex, it is possible that this may have an effect on the measured ductility of nanocrystalline materials. An important future experiment will be to subject nanocrystalline materials such as Cu or Al-Al_3Zr to disk-bend tests to compare results from disk-bend tests with those from tensile or compression tests.

The copper compression test results are consistent with those of Suryanarayanan *et al.* [12], who used compression testing to characterize somewhat larger grain-sized samples prepared by a solution-phase chemical processing route. In both studies, larger yield stresses and greater ductilities were observed compared to those observed in tensile tests of nanocrystalline Cu [6-10]. Suryanarayanan *et al.* [12] attribute the higher yield strengths they observed to the fact that their samples contained boron, possibly in the form of precipitates. An alternative explanation for the strengthening is that higher yield strengths are expected with compression testing than with tensile testing, as described previously. By using the gas condensation process for sample preparation in the present case, the formation of precipitates that occurred due to impurities in [12] was avoided and thus any ambiguities regarding the effects of precipitates on strengthening were eliminated. Even for recent tensile tests, where improvements in synthesis techniques led to minimization of the number of flaws present [9, 10], significantly lower yield strengths and strains-to-failure were observed than for the present compression tested samples. Since both the present compression samples and the earlier tensile samples [9, 10] were prepared in the same apparatus by the same techniques, it is apparent that flaws still play a major role in determination of measured mechanical properties of nanocrystalline materials.

In summary, the two main conclusions drawn from these studies are:

1) Materials that deform readily in coarse-grained form due to dislocation generation and motion become less ductile in nanocrystalline form since possible enhancements in diffusional deformation mechanisms are not sufficient to overcome competing reductions in dislocation-based deformation; on the other hand, for normally brittle coarse-grained materials in which dislocation-based deformation is already minimal, enhancements in diffusional-based deformation mechanisms may be sufficient to result in a net increase in ductility with decreasing grain size.

2) Despite recent improvements in synthesis techniques, the presence of flaws continues to have a substantial effect on the mechanical properties of nanocrystalline materials.

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