Supplemental Proceedings: Volume 1: Materials Processing and Interfaces
TMS (The Minerals, Metals & Materials Society), 2012

PROCESSING CHALLENGES OF DUAL-MATRIX CARBON NANOTUBE ALUMINUM COMPOSITES

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Keywords: Carbon Nanotube reinforced composites, Aluminum-CNT dual matrix composites

Abstract

The interest in nanostructured materials has grown considerably in recent years. Significant enhancements in strength have been reported. A common problem, however, is the associated reduction in the materials ductility. Efforts to overcome this ductility challenge by designing multi-modal or hierarchical microstructures have recently been reported. In this work, we report on the processing of dual matrix carbon nanotubes composites in which composite particles of aluminum reinforced with carbon nanotubes are embedded within a soft aluminum matrix. Such approach aims at combining the high strength of the CNT-reinforced region with the ductility of the soft aluminum matrix. Preliminary results, however, show that the interface quality is pivotal in promoting bonding between the two dissimilar particles since a poor interfacial bond is found to lead to deterioration in the composite properties; thus making it impossible to achieve the desired paradox of strength and ductility. Processing-related challenges in this regard are discussed.

Introduction

Due to their exceptional properties, carbon nanotubes (CNTs) have been recently used to reinforce materials in order to enhance their mechanical, electrical and thermal properties. The interest in CNT- metallic composites has been growing significantly over the past decade. In particular, carbon nanotube reinforced metal composites have been under intense investigations with the goal of generating composite materials with enhanced properties [1-3]. The consolidation of CNT-aluminum powder-based composites has so far involved processes such as hot pressing, hot extrusion, powder rolling, high pressure torsion, cold spraying, spark plasma sintering (SPS), and spark plasma extrusion (SPE) [4-13]. Most work has focused on the generation of composite materials with homogeneously dispersed carbon nanotubes. Nearly all the research groups reported enhanced mechanical properties to various degrees but this was generally combined with the undesirable loss in ductility.

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The dual matrix composite microstructural design, where the matrix is selectively reinforced in localized regions within the microstructure separated by ductile unreinforced matrix, may present significant benefits. This unique microstructural design has been documented to enhance both the wear resistance and toughness over conventional (single matrix) composites [14-16]. Other potential benefits include an ability to tailor the properties of the final composite which may give rise to controlled properties and enhanced formability.

A recent paper by two of the authors addressed the spark extrusion of single and dual matrix CNT-aluminum composites [17]. This paper presented for the first time CNT-aluminum composites based on the dual matrix design. However, the paper focused on the effect of the dual matrix design on the formability of the composite using spark plasma extrusion. No investigation of either the tensile behaviour of this novel material or its fracture toughness behavior was carried out. Furthermore, it has not yet been attempted to prepare the dual matrix composite using more conventional techniques such as compaction and hot extrusion; expected to provide more flexibility in preparing the unique sample geometry needed for fracture toughness tests.

Prior to adding the ductile matrix, the CNTs have to be well dispersed in the aluminum powders. As reported in our earlier publications, high energy ball milling has proven its effectiveness in dispersing the CNTs within the aluminum powders [7,18,19]. This is, however, accompanied by severe work hardening of the resulting composite powders as well as some coarsening in the powder particle size which necessitates controlling the amount of process control agent added. Such factors could result in processing challenges. Our current investigation addresses such effects by conducting preliminary research on the consolidation of these mixed powders in order to develop a better understanding of the impact of such differences on the feasibility of producing dual matrix composites as well as to identify conditions that lead to poor results so that they can be avoided.

Experimental Procedures

Al (99.7 % pure, – 200 mesh, Aluminum Powder Company Ltd., UK) and multi-wall carbon nanotubes (MWCNTs) (approximately 140 nm average diameter and 7 μm in length, supplied by the MER corporation, USA), were used in the present study. 5 wt.% CNT was used to prepare the composite powders with the balance being Al. The mixture was milled under argon in stainless steel jars using stainless steel milling balls (ball-to-powder ratio, BPR = 5:1) at 400 rpm for 30 min. The amount of methanol added as a process control agent (PCA) to reduce the particle welding was limited to 50μL. To prevent heat build-up during the milling process which would result in the formation of undesirable reaction products at the interface of CNTs and aluminum, the ball milling process was interrupted every 10 min. Milled composite powders were then added to equal weight of unmilled aluminum powder (milled for 5 min at 200 rpm in order to break off the oxide layer) and were turbula mixed for 30 min to produce dual matrix powders. Those were then compacted at 475 MPa. Hot extrusion of the compact was conducted at 500°C using an extrusion ratio of 4:1. Dog-bone tensile test samples were machined and super-finished out of the extrudates, and tested to fracture in order to generate a stress-strain curve.

Scanning electron microscopy (SEM) was used to determine the powder morphology following the milling process as well as investigating the fracture surface following the tension test in order

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to evaluate the interface strength. For microstructural characterization, extruded specimens were sectioned along the cross-section, ground and polished to 1 micron finish.

Results and Discussion

Initial efforts were focused on evaluating the particle size and morphology following the milling process as well as the dispersion of the CNTs. As can be seen from Figure 1, the particle size is not uniform; two distinct particle sizes are observed (18 μm and 75 μm). The presence of 5 wt% CNT favoured particle fracturing and thus a decrease in the particle size of the milled composite powders, as noted in our previous publication [19]. It is also noted that dispersed CNTs are present on the surface of the particles (Figure 1 b). Ideally, CNTs should be embedded within the particles as otherwise they could sustain damage under the impact of the milling media. Additionally, CNTs residing on the particle surface could interfere with subsequent particle bonding during the consolidation process. The addition of the PCA was necessary because trials in which the PCA was eliminated resulted in particle sticking to both the jars and the milling balls and thus low powder yield. Upon mixing the milled powders with equal amounts of unmilled aluminum powders the resulting morphology observed is presented in Figure 2.

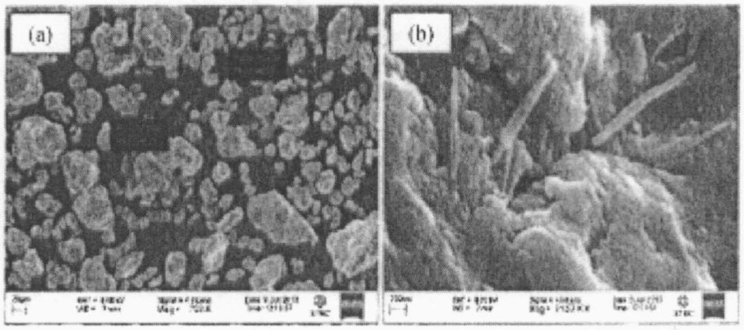

Figure 1. SEM micrograph of (a) milled Al-5wt%CNT composite particles and (b) a higher magnification image of one of the particles showing several CNTs.

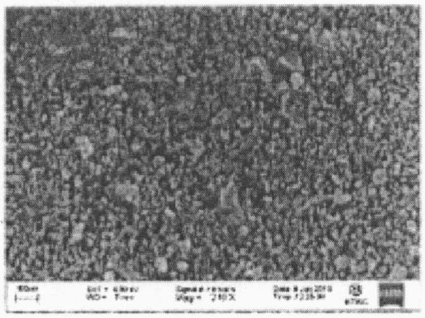

Figure 2. Dual powders (50% Al – 50% (Al-5wt% CNT)) mixed in the turbula mixer for 30 min.

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A representative stress-strain diagram of a dual matrix extruded sample is presented in Figure 3. A maximum strength of 165.5 MPa and a maximum strain of 1.3% (measured using a clip-on extensometer) represent the best results obtained. Such strength value which is 38% higher than the average value obtained for pure unmilled aluminum samples (typically 120 MPa), prepared under the same conditions, confirmed the reinforcing effect of the composite particles. However, this was accompanied with the undesirable effect of a severe loss in the ductility of the samples.

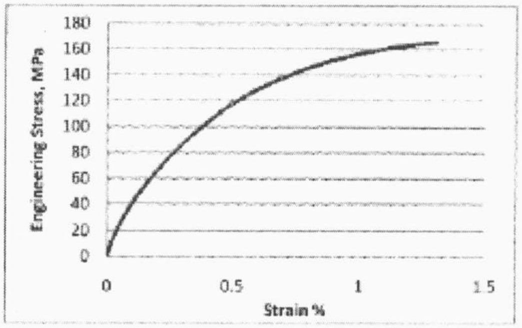

Figure 3. Stress strain diagram of a dual matrix (50% Al – 50% (Al-5wt% CNT)) sample.

Evaluation of the fracture surface by SEM revealed clearly (Figure 4 a) the difference in behavior between the soft unreinforced aluminum which developed large dimples as opposed to the heavily strain hardened milled composite particles which don't seem to have deformed as much. Figure 4 b is a higher magnification image which focuses on the interface between two particles and shows porosity along the interface due to the difference in deformation behavior between the two particles which appear to easily debond. The lack of a strong bond at the interface could also be attributed to the presence of some CNTs on the surface, as noted earlier, as well as other contaminants or oxide films. It is believed that the weak interface is responsible for inefficient load transfer from the outer soft aluminum matrix to the inner harder particles which lead to the observed unsatisfactory mechanical behavior.

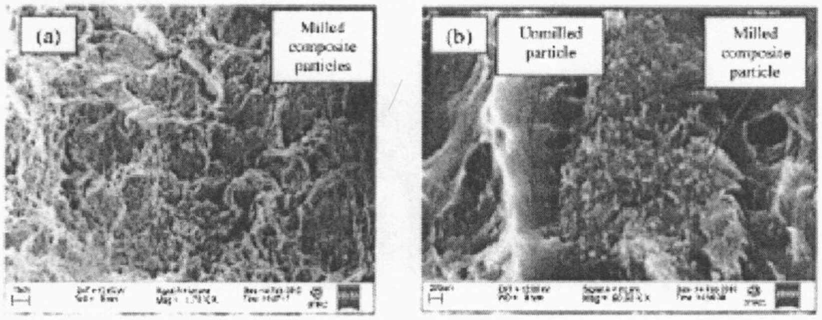

Figure 4. Fracture surfaces of a dual matrix extruded sample showing (a) the two distinct regions (b) the interface between an unreinforced particle and a strain-hardened composite one.

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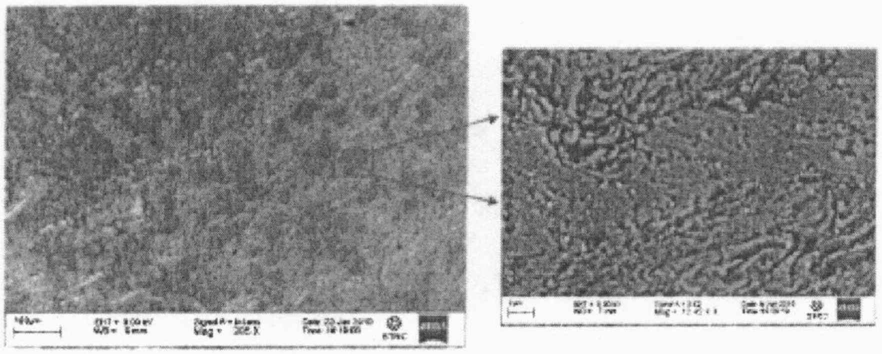

Figure 5. Microstructure of dual matrix composites showing the distribution of the CNT-Al composite particles (dark) within the aluminum outer region. The inset shows details of the CNT-Al composite particles showing pores as well as oxide particles.

Figure 5 shows the microstructure of polished and etched samples. The inset focuses on a composite particle and reveals many pores surrounding the CNTs. Such poor bond would also contribute to the overall inadequate mechanical behavior.

Recent investigations attempting to overcome the previously explained shortcomings addressed the problem of CNTs residing on the particle surface as well as particle coarsening and non-uniform size by milling the powders for 1 hr instead of 30 min using a BPR of 5:1, as well as the use of higher purity PCA. Careful handling of the milled powders (which were kept under an argon atmosphere throughout the process) was also exercised in order to minimize any oxidation of the surface-active powders following milling. SEM observations (not shown) confirmed that the modified milling conditions lead to CNTs being embedded within the particles as well as a more uniform particle size. The milled composite powders were then mixed with equal weight of unmilled powders by further milling for an additional 1 hour. The final powder morphology is shown in Figure 6 and shows the more uniform particle size. Homogenization time before extrusion was extended to 1 hr instead of 30 min to ensure better consolidation.

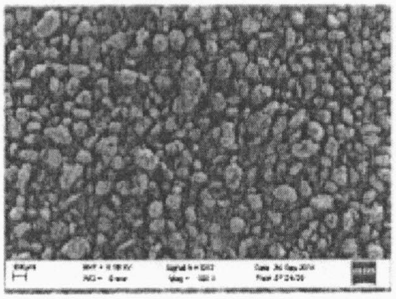

Figure 6. Dual powders (50% Al – 50% (Al-5wt% CNT)) milled for an additional 1 hr.

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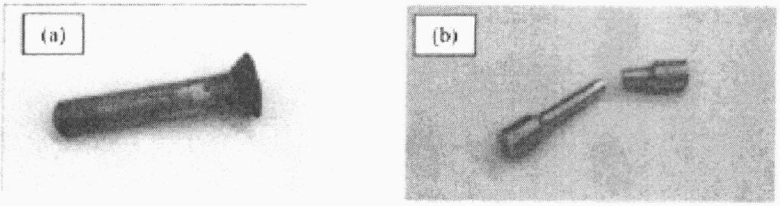

Figure 7. (a) Extruded dual matrix sample (b) Broken tensile test sample.

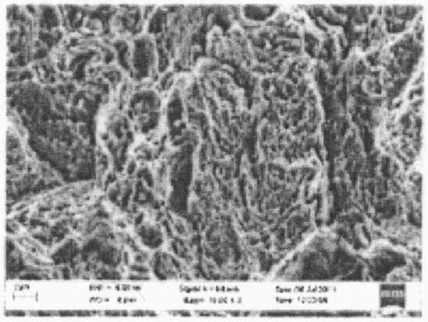

Figure 8. SEM micrograph of the fracture surface of a dual matrix extruded sample.

Preliminary results show a noticeable increase in the formability of the samples. The extruded dual matrix composites were free of extrusion defects at the surface (Figure 7 a), as opposed to the earlier samples in which surface cracks in the form of bamboo defects were frequently observed. In addition, noticeable enhanced ductility with consistent failure in the gauge length was observed (Figure 7 b). The initial results of the tension samples show much improved strength reaching 365 MPa as well as enhanced ductility reaching 6.7%. Fracture surface investigations (Figure 8) reveal overall ductility and less noticeable difference in the dimple size and depth between the two regions as opposed to the earlier samples in which ductility was only observed in the unmilled regions. This research is still the subject of ongoing work, but the results are encouraging and it is expected that this novel microstructural design would result in composites with reasonable ductility in addition to the enhanced mechanical properties. Samples with other CNT contents as well as different ratios of unmilled versus composite powders will also be prepared and evaluated.

Conclusions

A number of recommendations can be made regarding the dual matrix samples:

1. The dual matrix design has the potential to produce samples of high strength and good ductility.
2. Controlling the milling conditions so as to yield fine uniform particles as well as embedded CNTs is desirable.

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3. Ensuring a clean metallurgical interface by careful handling of the milled powders to minimize oxidation is essential.

Further work is underway to compare the dual matrix composites to single matrix ones processed under the same conditions. Preparation of fracture samples that allow us to characterize the fracture behavior is also in progress.

Acknowledgments

The authors wish to acknowledge the financial support of the US-Egypt Joint Science and Technology fund (grant number MAN11-011-007) and the National Science Foundation (Office of International Science and Engineering) under grant number 0710869, the Yousef Jameel Science and Technology Research Center (YJSTRC) and the office of the Associate Provost for Research Administration at the American University in Cairo, Egypt.

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