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Constitutive Characterization of Multi-Constituent Particulate Composites

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Multi-constituent epoxy-based particulate composites consisting of individual particles of aluminum and a second phase (copper, nickel or tungsten) have been synthesized. The mechanical and physical properties of the composite depend on the mechanical and physical properties of the individual components; their loading density; the shape and size of the particles; the interfacial adhesion; residual stresses; and matrix porosity. These multi-phase particulate composites have been generated to investigate the deformation of aluminum in the presence of the second phase. Quasi-static and dynamic compression experiments have been performed to characterize the materials. The microstructures of the quasi-statically and dynamically deformed samples have been quantified to determine the amount of deformation in the aluminum particles, as a function of their proximity (i.e. near or far) from the second phase particles.

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Constitutive Characterization of Multi-Constituent Particulate Composites

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Abstract
Multi-constituent epoxy-based particulate composites consisting of individual particles of aluminum and a second phase (copper, nickel or tungsten) have been synthesized. The mechanical and physical properties of the composite depend on the mechanical and physical properties of the individual components; their loading density; the shape and size of the particles; the interfacial adhesion; residual stresses; and matrix porosity. These multi-phase particulate composites have been generated to investigate the deformation of aluminum in the presence of the second phase. Quasi-static and dynamic compression experiments have been performed to characterize the materials. The microstructures of the quasi-statically and dynamically deformed samples have been quantified to determine the amount of deformation in the aluminum particles, as a function of their proximity (i.e. near or far) from the second phase particles.

Introduction
Particulate composite materials composed of one or more varieties of particles in a polymer binder are widely used in military and civilian applications. They can be tailored for desired mechanical properties with appropriate choices of materials, particle sizes and loading densities. Several studies on similar epoxy-based composites have been reported and have shown that particle size [1,2], shape [3], and concentration [4] and properties of the constituents can affect the properties of particulate composites. In composites of Al_2O_3 particles in epoxy, increasing the particle concentration and decreasing the particle size were found to increase the stress at 4% strain [5]. A study of aluminum-filled epoxy found adding a small amount of filler (~ 5 vol.%) increased the compressive yield stress, but additional amounts of filler decreased the compressive yield stress [6]. However, tests on glass bead/epoxy composites found that increasing the volume percent of glass bead filler increased the yield stress and fracture toughness of the material [7,8].

Several multi-phase particulate composites have been generated to investigate the deformation of aluminum particles in the presence of a second metallic phase. In this paper, single phase (aluminum and epoxy) and multi-phase (aluminum-metal-epoxy, where metal is copper, nickel, or tungsten) have been prepared. The samples have been deformed at quasi-static and dynamic strain rates and the deformed microstructures have been examined to determine the strain in the particulates.

Experimental Procedure
Five materials were prepared for this study – two composites containing only aluminum and epoxy, with two different volume fractions of aluminum, and three composites containing an additional second metallic phase, at a fixed volume fraction. The manufacturer and average particle size for the powders are given in Table 1. The appropriate volume fractions of powder for each composite were blended into Epon 826 and cured with diethanolamine (DEA). The composite mixture was cast into blocks and appropriate samples were machined. The density of each composite was measured using pyncnometry. The sample names with corresponding volume fractions of metal powders and the measured density are reported in Table 2.

Compression experiments at quasi-static strain rates were conducted with an MTS 810 testing system with a 100 KN test frame. Care was taken to center the samples on the platens prior to testing. MTS software was used to conduct constant displacement rates tests at a strain rate of $9.4 \times 10^{-4} s^{-1}$. A thin layer of PTFE tape was used to lubricate the surfaces of the platen in contact with the test specimen. It was found that this provided better lubrication than a film of Boron Nitride (BN) with a layer of Molybdenum disilicide ($MOSi_2$) on top that was used in

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previous studies [9]. In addition to the MTS system recording the loads and displacement of the frame, interfacing software between the test frame and a video extensometer system (VIC Gauge 2.0 from Correlated Solutions

Table 1: Precursor powder characteristics

Powder	Supplier	Average Particle Size (μm)
Aluminum (X81)	Toyal	27
Copper	Atlantic Equipment Engineers	37
Nickel	Atlantic Equipment Engineers	44
Tungsten	H.C. Starck (Kulite)	37

Table 2: Material compositions and measured densities used in this study.

Material	Density (g/cm^3)	Al Vol%	Cu Vol%	Ni Vol%	W Vol%	Epoxy Vol%
Epoxy-35Al	1.725	35				65
Epoxy-45Al	1.875	45				55
Epoxy-Al-Cu	2.475	35	10			
Epoxy-Al-Ni	2.513	35		10		
Epoxy-Al-W	3.652	35			10	

Inc.) read input voltages for both the load and displacement. Additionally, this software interfaces with a video system, which allows the user to place virtual displacement gages on the specimen that are tracked as testing takes place. Multiple virtual displacement gages were used for comparison and to enable the test to continue in the event that one gage failed. Samples were loaded to, nominally, 10%, 20% and 30% strain. The samples were then used for post-mortem analysis.

Compression experiments at intermediate strain rates (approximately $1x10^3$ and $5x10^3$ s^{-1}) were conducted using a split Hopkinson pressure bar (SHPB) system [10]. The experiments were conducted using the SHPB system located at AFRL/RWME, Eglin AFB, FL, which is comprised of 1524 mm long, 12.7 mm diameter incident and transmitted bars of 6061-T6 aluminum. The striker is 610 mm long and made of the same material as the other bars. The samples, which were nominally 8 mm diameter by 3.5 mm thick and 5 mm diameter by 2.5 mm thick, are positioned between the incident and transmitted bars. The bar faces were lightly lubricated with grease to reduce friction.

After quasi-static or dynamic testing, representative samples of each material were sliced along the centerline of the specimen, such that a longitudinal section containing the loading direction was visible. This face was mounted, polished, and examined using Scanning Electron Microscopy (SEM). In order to ensure statistical rigor, several images of each sample were obtained, thereby providing metallographic sections of ~100 particles for each of the conditions that were analyzed – (i) aluminum particles in close proximity to second phase particles; (ii) aluminum particles positioned away from second phase particles and; (iii) second phase particles positioned away from aluminum particles.

A deforming particle can be used as a local strain gauge. Assuming that the particle volume is conserved during deformation and that a spheroidal particle with an initial aspect ratio of 1 will deform as an oblate spheroid with its minor axis oriented along the deformation axis in the material, then any longitudinal 2-D section through the particle will have an aspect ratio, γ, directly related to the particle strain, $\bar{\varepsilon}$, by

$$\bar{\varepsilon} = -\frac{2}{3} ln(\bar{\gamma}). \quad (1)$$

In single phase samples, the aspect ratios of ~100 aluminum particles were measured to determine the average strain. In the multi-phase particulate composites, aluminum particles positioned “near” to second phase particles – i.e. those aluminum particles which had a second phase particle as a nearest neighbor – were measured along with aluminum particles that were positioned “far” from any second phase particle, i.e. those that had several particle diameters between themselves and the second phase. Additionally, the aspect ratios of the second phase particles (copper, nickel, or tungsten) were also measured.

Results and Discussion

Stress-strain curves from the five composites that were studied are presented in Figure 1. There is very little difference in the stress-strain responses of the different composites. Since the stress-strain behavior is generally dependant on the volume fraction of particles, small variation is expected in these materials having comparable volume fractions. At strains above 0.05, each of the multi-phase composites show higher flow stresses than the aluminum-containing composites, which rank according to volume fraction of aluminum. At strains below 0.05, The lower volume fraction aluminum composite (Epoxy-35Al) appears to have higher strength than the higher volume fraction composite (Epoxy-45Al) in the quasi-static regime. White, et al. have shown the presence of a percolation threshold in similar composites at a similar level of loading [11], which may account for this difference. This discrepancy is not seen in the dynamic data, where yield and flow stresses all rank according to volume fraction and presence of 2^{nd}-phase particles.

The primary focus of this work is analysis of the strain measured in the particles themselves compared with the global strain measured on the sample. For the quasi-static experiments, these measurements were taken at three levels of strain (0.1, 0.2, and 0.3) and the results are shown in Figure 2. For the dynamic experiments, the level of strain is a result of the sample dimensions and is not as controllable as in the quasi-static experiments, but nevertheless ranges from ~0.3 – 0.45. The results from the dynamic experiments are given in Figure 3.

Figures 2 (a) and 3 (a) compare the strain in aluminum particles positioned "far" from second phase particles in the multi-phase composites and the strain measured in the aluminum particles in the aluminum-epoxy composites, for the quasi-static and dynamic experiments, respectively. In the quasi-static experiments, where the global true strain is precisely controlled, the strain in the aluminum particles tends to cluster above the global true strain, indicating efficient load transfer between the epoxy matrix and the stiffer reinforcement. In addition, the strains measured in the aluminum particles in the multi-phase composites tend to be lower than the strains in the aluminum composites without the second-phase particles, suggesting a stiffening of the matrix (effectively shielding the aluminum particles) by the addition of the second phase. This is consistent with the trends for flow stress shown in Figure 1. In the dynamic experiments, the strain in the aluminum-epoxy composites and in the aluminum particles far from the second phase seems to compare with the global true strain in the sample indicating that these particles are deforming with the epoxy matrix in a homogeneous manner. The difference between the quasi-static experiments and the dynamic experiments may result from load transfer across the

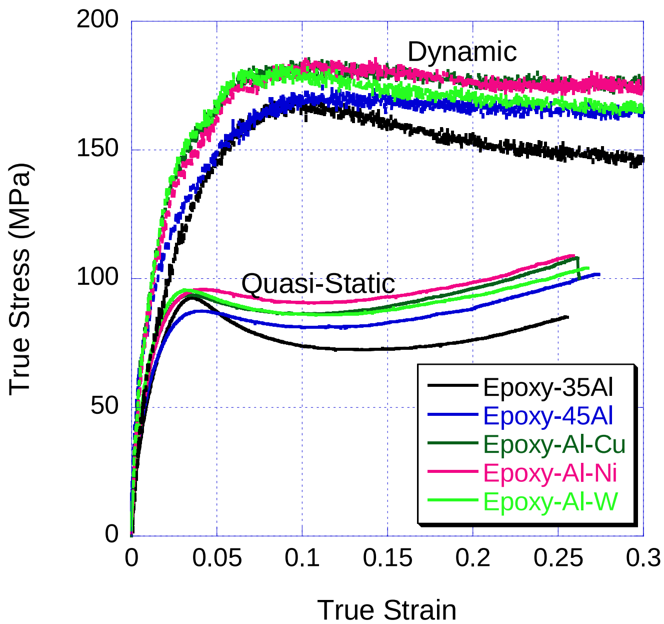


Figure 1: Stress-strain curves for each composite at quasi-static (9×10^{-4} /s) and dynamic (~1.7×10^{3} /s) strain rates.

interface between matrix and particle, which may be less efficient at higher loading rates, leading to lower overall strains in the particles. Moreover, the increased dynamic stiffness of the epoxy matrix may play a key role in reducing the apparent load partitioning between the matrix and reinforcement. In this case, the addition of the stiffer tungsten particles would have a greater effect than either the addition of copper or nickel, which is observed.

In both the quasi-static and dynamic experiments, the aluminum particles positioned “near” to second phase particles showed increased strain over those particles that were positioned “far” from the second phase particles, as shown in Figures 2 (b) and 3 (b). In the quasi-static experiments, where measurements were made at different levels of strain, the strains in the aluminum particles near to the second phase particles are consistently higher than the strains measured in aluminum particles far from the second phase particles, *at all strain levels.* In every case, the aluminum particles strain to a greater extent than predicted by the global true strain, indicating efficient load transfer between matrix and particle. However, the measured strains in the second phase particles appear to be independent of the global strain imposed on the composite. This may suggest that the stiffer (and stronger) second phase particles do not deform as readily as the aluminum particles, and simply move as rigid-bodies while the epoxy matrix and the aluminum particles undergo deformation. The enhanced deformation in the aluminum particles in close proximity to second phase particles suggests that the second phase particles act as either hammers or anvils in encouraging the aluminum particles to deform. Clearly, in spatially-heterogeneous composite microstructures such as these, local effects of microstructure, including locally high volume fractions of second phase particles would be expected to play an active role during deformation, beyond simply stiffening the epoxy matrix. In the dynamic loading regime, the strains measured in aluminum particles near to second phase particles are again consistently higher than strains measured in aluminum particles far from second phase particles, although the overall levels of strain are reduced, consistent with the load-sharing arguments presented above. In all cases, the second phase particles show the lowest strains, however, there is a clearer trend of increasing particle strain with increasing global true strain than was observed in the quasi-static data. This may indicate that the dynamically-stiffened matrix imparts sufficient load to the second phase particles to get them to deform. However, it should also be noted that even at the lowest applied strains, under quasi-static loading, there is an apparent ‘residual strain’ in the second phase particles, between 0.15 – 0.20. This may indicate an initial ‘non-sphericity’ of the particles, in the starting powders, which translates into systematic error in the strain measurements at all strain levels. Further examination of the starting powders and/or measurements on undeformed specimens are necessary in order to rule out this effect.

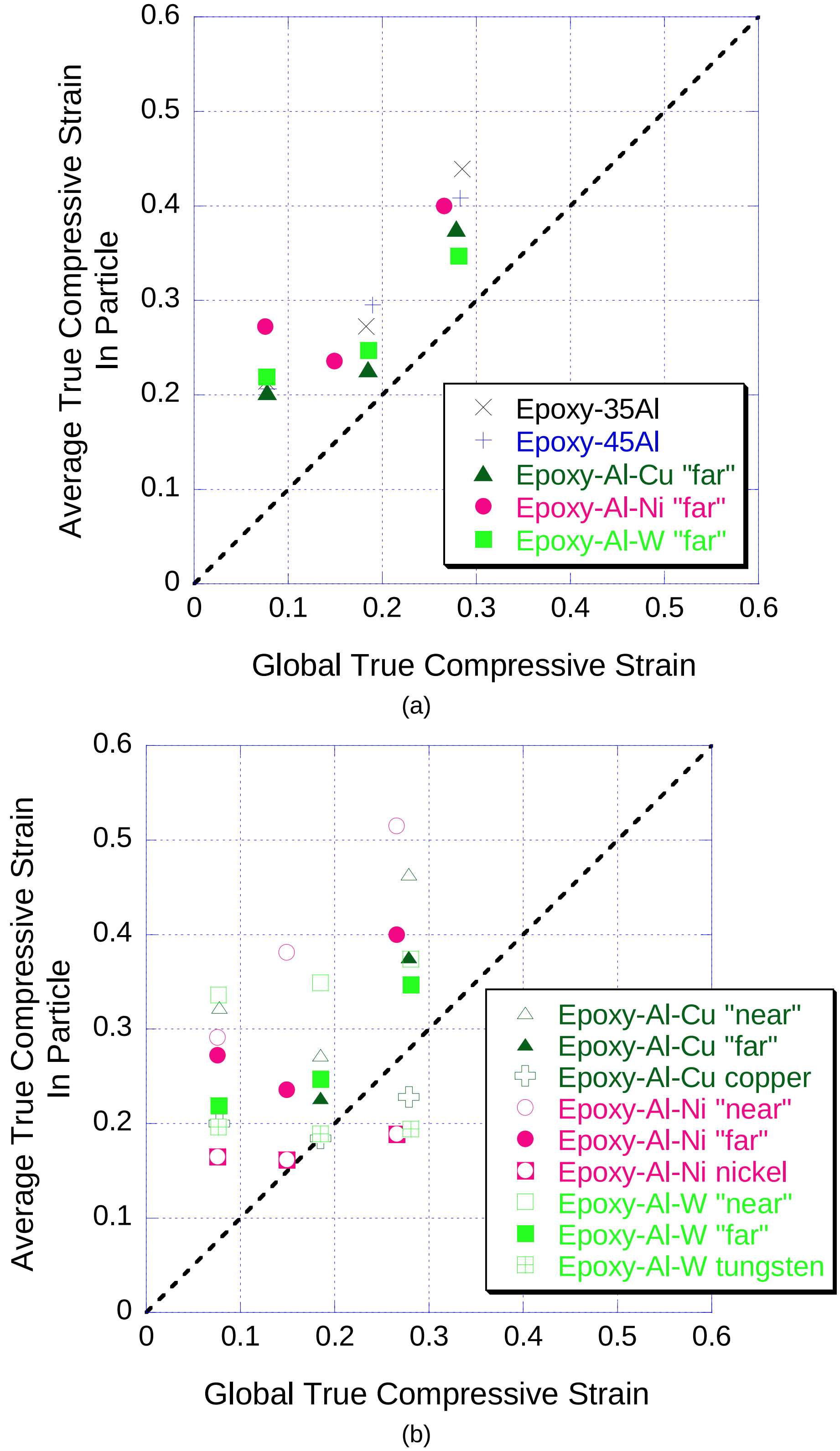
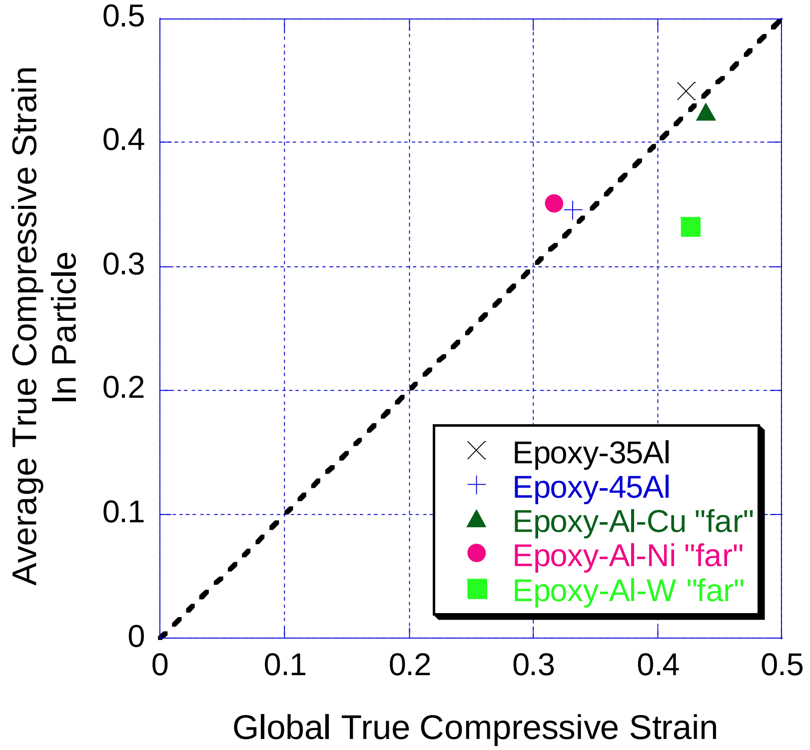
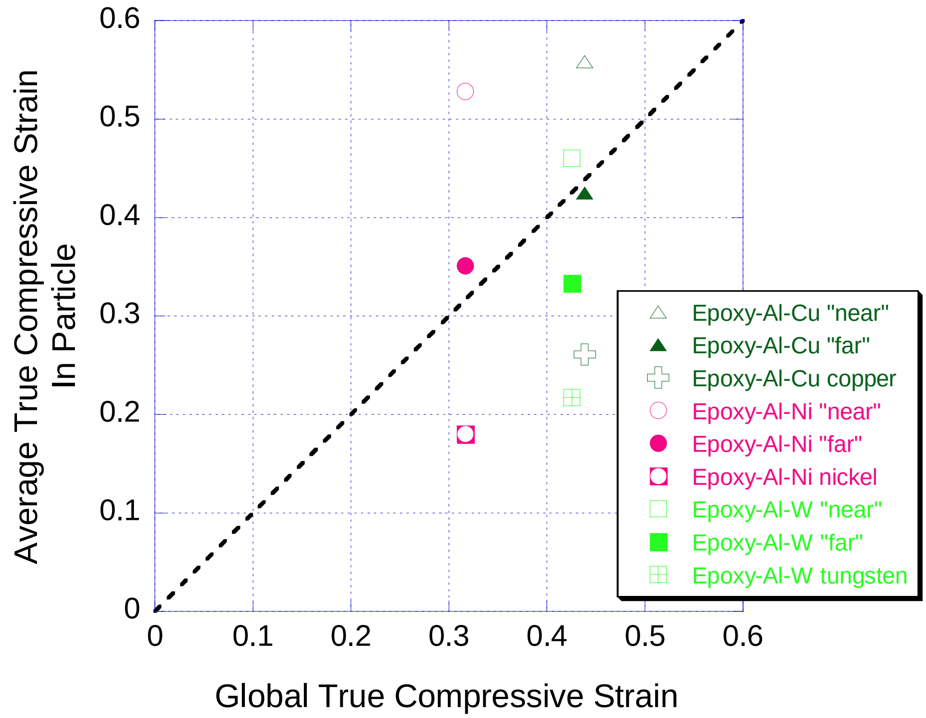


Figure 2: Quasi-static global true compressive strain versus average true compressive strain for (a) single phase (Al only) particulate composites and two-phase particulate composites with Al particles far from the second phase and (b) two-phase particulate composites, with Al and Cu, Ni or W particulates, where near indicates Al particles close to the second phase, far indicates Al particles far from the second phase.



(a)



(b)

Figure 3: Dynamic global true compressive strain versus average true compressive strain for (a) single phase (Al only) particulate composites and two-phase particulate composites with Al particles far from the second phase and (b) two-phase particulate composites, with Al and Cu, Ni or W particulates, where near indicates Al particles close to the second phase, far indicates Al particles far from the second phase.

Summary

Multi-constituent epoxy-based particulate composites consisting of individual particles of aluminum and a second phase (copper, nickel or tungsten) have been synthesized to investigate the deformation of aluminum in the presence of the second phase. Quasi-static and dynamic compression experiments have been performed to characterize the materials. The microstructures of the quasi-statically and dynamically deformed samples have been quantified to determine the amount of deformation in the aluminum particles, as a function of their proximity to the second phase particles. In both the quasi-static and dynamic experiments, the aluminum particles that were close to the second phase particles showed increased strain over those that were far from the second phase particles. Furthermore, decreased load partitioning between matrix and particle was observed in the dynamic experiments.

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Opinions, interpretations, conclusions and recommendations are those of the authors and are not necessarily endorsed by the United States Air Force.

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