STUDY OF INTERFACES IN XD™ Al/TiC_p METAL MATRIX COMPOSITES

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ABSTRACT

The metal-ceramic interface in an XD™ Al/TiC_p metal matrix composite has been characterized in as-extruded, recrystallized, and high temperature heat-treated conditions. In both the as-extruded and recrystallized composite, the interface is atomically abrupt. Localized orientation relationships exist between Al and TiC that lead to some degree of coherency at the interface. Recrystallization produces semicoherent interfaces by formation of subgrains in the Al adjacent to the TiC particles. Few interfaces show cracking, even after extensive deformation. Lack of cracking together with the direct contact down to atomic level, observed between the two phases are evidence for excellent bonding between the carbide particles and the aluminum matrix. Heat treating samples at 913 K for 24 hours produces reaction products like Al_3Ti and Al_4C_3. These reactions are explained on the basis of thermodynamic data.

INTRODUCTION

In recent years there have been many studies on solid state metal-ceramic interfaces, with emphasis on structure and chemistry on an atomic scale [1-2]. Very little, however, is known about the structural aspects of specific interfaces in particulate metal-matrix composites. It is a well known fact that the interfaces play an important role in the performance of composites. The nature of the bonding and the interfacial energy are functions of the structure and chemistry of the interfaces on an atomic scale. Thermodynamic stability of the interface also is of interest. The processing parameters of course influence the nature of the interface obtained in commercial composites.

This paper reports a study of the metal-particulate interfaces in an Al/TiC_p composite prepared at Martin Marietta Laboratories, Baltimore, Maryland, by the XD™ process [3]. Both Al and TiC have a fcc lattice. TiC is partly ionic [4] and has a NaCl structure. The present work is a qualitative study of the structure observed at the interface in order to understand the nature of the interfacial bonding. The thermodynamic stability of the interface has also been examined.

MATERIAL

Aluminum with 15 volume percent TiC, of 0.7 µm average particle diameter, was produced by a two step process. First, TiC was precipitated *in situ* in molten Al via the XD™ process; this step produced a high volume fraction "master alloy". In the second step, the XD™ master alloy was diluted with 99.99% pure aluminum and cast into a 75 mm diameter mold. This ingot was then extruded at 648 K with a 27:1 extrusion ratio. The TiC particles

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produced are single crystals with surfaces free of impurities.

EXPERIMENT

TEM samples were prepared by mechanical polishing followed by argon ion thinning using a liquid nitrogen-cooled holder. The microstructure of an as-extruded sample was studied by conventional transmission electron microscopy (CTEM) on a Hitachi H700-H TEM. High resolution transmission electron microscopy (HRTEM) was carried out on a Hitachi H9000 TEM. The H9000 microscope was operated at 300 kV and images were obtained without an objective aperture. All the images were recorded at an optimum defocus value, where maximum details could be seen of the interface and of the Al and TiC lattice.

Some of the as-extruded composite was cold rolled to 75% reduction and annealed for recrystallization at 773 K for 1 hour. Samples of the recrystallized composite were studied using CTEM and HRTEM to investigate the interface between newly formed grains and the particles.

Samples of the as-extruded composite were heat treated at 913 K (20 K below the melting point of Al) for 24 hours to investigate the thermodynamic stability of the interface. The interfaces in these heat-treated samples, also were examined by CTEM and the observations explained on the basis of thermodynamic considerations.

RESULTS AND DISCUSSION

The as-extruded composite has a very uniform distribution of particles. Figure 1 shows a TEM micrograph of an equiaxed Al grain with TiC particles. TiC particles are found at Al grain boundaries as well as inside the grain.

Interface Structure

Figure 2 is a TEM micrograph of an Al/TiC interface in an as-extruded sample. It shows a regular array of facets and dislocations, which are indicative of some degree of semi-coherency at the interface. Similar arrangements were seen at some other interfaces, as discussed below.

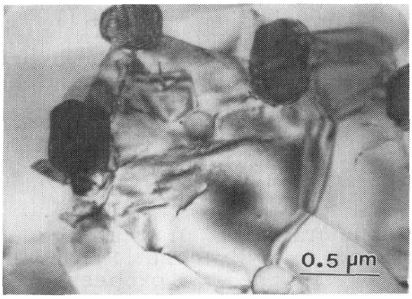


Figure 1. TEM micrograph of an Al grain with TiC particles in an as-extruded sample.

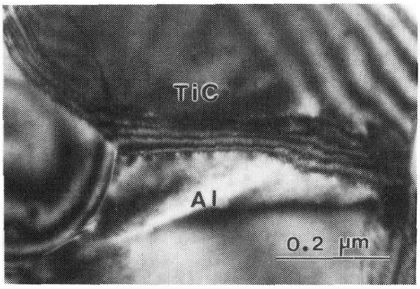


Figure 2. TEM micrograph of an Al/TiC interface showing facets and dislocations in as-extruded sample.

Figure 3 shows the lattice image of an Al/TiC interface in the as-extruded sample, recorded with the beam parallel to the [011] zone axis of TiC. Al is slightly off [011] zone axis orientation. The interface is atomically abrupt and shows regions of good matching, where every alternate $(\bar{1}1\bar{1})$ plane of TiC appears to be connected to a $(\bar{1}1\bar{1})$ plane of Al. There is a local orientation relationship between Al and TiC that leads to this kind of periodicity. Examination of many other interfaces shows that no general orientation relationship exists between Al and TiC. However in some cases there is a tendency to form special boundaries. These might have formed during solidification by nucleation of Al grains at TiC sites or by partial recrystallization during hot extrusion. Both scenarios are possible since TiC particles are formed by an *in-situ* reaction and have a clean surface. So for further studies, interfaces were studied after recrystallization.

In the recrystallized samples, the Al grains contain subgrains near each of the TiC particles. The subgrains are oriented to have a low energy interface with the particle. Figures 4(a) and (b) are bright and dark field micrographs that show the interface between an Al subgrain and TiC. Strain contrast and dislocations can be seen at regular intervals. The dislocations probably occur to reduce the misfit. Such formation of subgrains has been observed previously at diffusion-bonded Nb/Al_2O_3 interfaces. The subgrains reduce the energy due to lattice mismatch [2]. Dislocations have been seen at inclined Al/TiC interfaces over a range of relative orientations. An

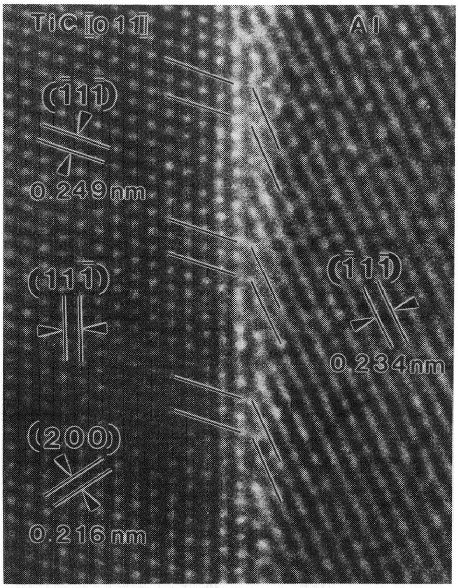
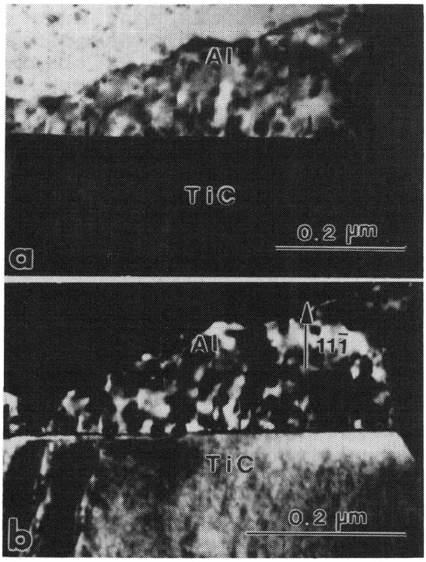
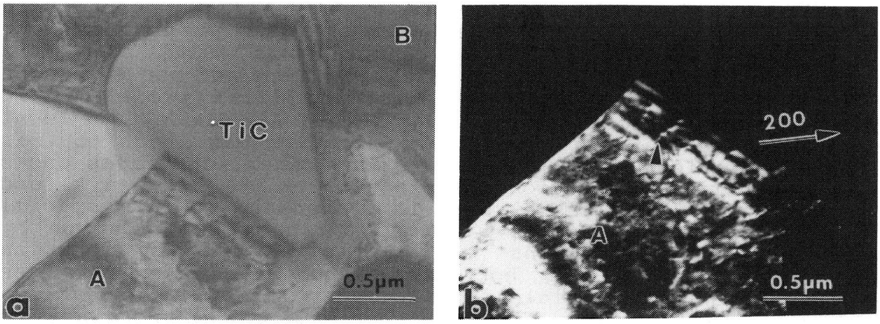


Figure 3. HRTEM micrograph of an Al/TiC interface in as-extruded sample showing an atomically abrupt interface. Regions of good matching shown.



Figures 4(a)-(b). Bright and dark field TEM micrographs of an interface between a TiC particle and an Al subgrain in recrystallized sample. Strain contrast and dislocations can be seen inside the Al at the interface.



Figures 5(a). A TiC particle surrounded by Al subgrains, marked 'A' and 'B'. Dislocations are observed at the inclined interfaces. (b) Dark field micrograph of subgrain 'A', showing dislocations (arrowed) at the boundary with the particle.

example is shown in figures 5(a) and (b). These observations prove that some degree of coherency exists at interfaces between Al subgrains and TiC.

Figure 6(a) is an interface between an Al subgrain and TiC from the recrystallized sample. The interface is highly faceted. Figure 6(b) is a magnified view of the area marked in Figure 6(a). The TiC is close to the [011] zone-axis orientation. Al shows only one dimensional fringes representing (111) planes as it is not in the zone axis orientation. It is clear that the interface is highly faceted even on an atomic scale and almost perfect matching can be observed across the interface. This kind of local coherency has been seen in many other interfaces, where it was possible to tilt the TEM sample to observe the interfaces 'edge-on'.

It is speculated that during recrystallization, the newly formed Al grains tend to orient themselves near TiC particles so as to minimize the interfacial energy and increase the degree of coherency. The misorientation between the particle and the matrix differs from particle to particle and around the same particle.

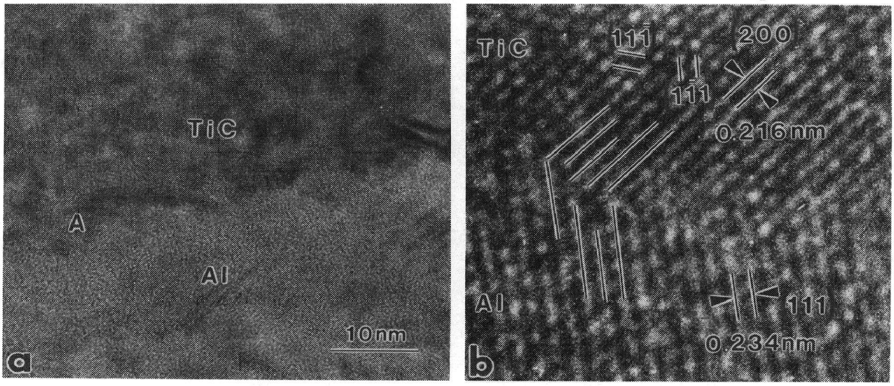


Figure 6(a). A low magnification micrograph of highly faceted interface in recrystallized sample. (b) Enlargement of part 'A' of Figure 6(a) showing facetting on atomic scale and large degree of lattice matching.

This happens since the same grain of Al can have many differently oriented particles, as shown in Figure 1. Hence it is not possible to get the same degree of coherency at all the interfaces. However, a large degree of local coherency has been observed, facilitated by formation of small angle boundaries near interfaces, faceting and dislocations at the interface. These mechanisms lower the interfacial energy. Observation of special boundaries in the as-extruded composite is possibly due to the effect of recrystallization of Al grains at TiC sites during the hot extrusion process. The chances of a naturally occurring low energy interface are increased by the fact that the surfaces of TiC, which is precipitated *in-situ*, are without impurities. This leads to a kind of chemical bonding with direct interaction between the two phases on an electronic scale [5]. TEM examination of the heavily cold-rolled sample showed voids only near clusters of particles. Otherwise, voids were not seen in as-extruded or cold-rolled and recrystallized samples. This observation suggests that the interface is ductile and that the bonding is metallic in nature.

Thermodynamic Stability

To determine the thermodynamic stability of the interface, the composite was heat treated at 913 K for 24 hours. This prolonged heating results in the formation of a reaction zone of Al_4C_3 and Al_3Ti at the interface between Al and TiC. Such a reaction zone is shown in Figure 7. The identity of the reaction products has been determined by selected area diffraction and dark-field techniques.

To explain the reaction zone, three equations can be considered :

$$3\ TiC + 13\ Al\ (l/s) \longrightarrow Al_4C_3 + 3\ Al_3Ti \quad (1)$$
$$3\ TiC + 7\ Al\ (l/s) \longrightarrow Al_4C_3 + 3\ TiAl \quad (2)$$
$$3\ TiC + 4\ Al\ (l/s) \longrightarrow Al_4C_3 + 3\ Ti \quad (3)$$

Figure 8 shows free energy change versus temperature curves [6] for all three reactions. It is clear that reaction (1) is expected to occur below 1025 K, because the free energy change is negative. Reactions (2) and (3) have a positive free energy

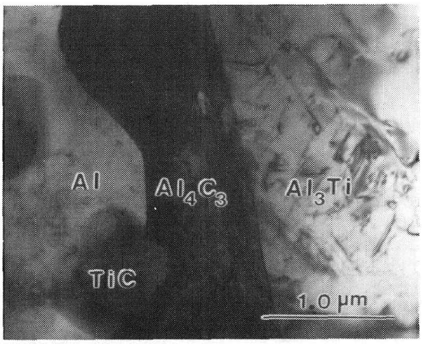


Figure 7. TEM micrograph showing Al_4C_3 and Al_3Ti formed at Al/TiC interface after heat treating.

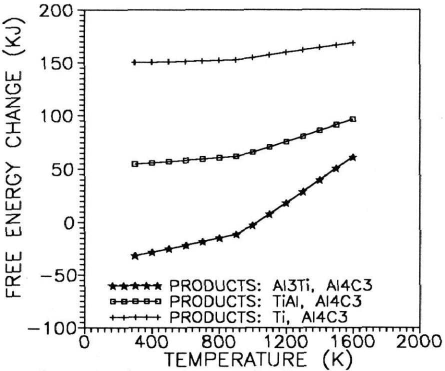


Figure 8. Plot of free energy change versus temperature for interfacial reactions.

change at all temperatures of interest and hence are not feasible. These curves explain the formation of reaction products at Al/TiC interfaces.

The tendency to form reaction products at the interface indicates that careful monitoring of temperature and time is necessary for processing. The interfaces in as-received samples were atomically sharp. No reaction product could be seen. No changes were observed on holding for 168 hours at 773 K. This is because of the very slow diffusion kinetics at that temperature. Thus a reaction product can be avoided by processing the composite above 1025 K and controlling the cooling rate or using sufficiently low temperatures for solid state processing.

CONCLUSION

The interfaces in as-extruded, recrystallized and high temperature heat treated XD™ Al/TiC composite have been studied using conventional and high resolution microscopy techniques. The interfaces are atomically abrupt in the as-extruded and recrystallized samples. The occurrence of special boundaries appears to become more frequent on recrystallization as Al subgrains form adjacent to the TiC particles so as to produce some degree of interfacial coherency. The semicoherency is achieved by local orientation relationships, dislocations and by faceting. Such a high degree of coherency at local levels illustrates the tendency of a metal-ceramic interface in an *in-situ* composite to reduce its energy [5]. This reduction is possible because the TiC particles are free of impurities. The reduction process leads to good bonding at the interface.

The interface obtained after 24 hours of heat treatment at 913 K shows formation of the reaction products Al_3Ti and Al_4C_3. The reaction leading to these products is thermodynamically favorable below 1025 K. However, it does not take place at an observable rate at temperatures like 773 K. Hence, precautions can be taken in choosing the processing temperatures of this metal-matrix composite to obtain a sharp interface.

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