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Aluminum/diamond composites: A preparative method to characterize reactivity and selectivity at the interface

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Reactivity at the interface in aluminum/diamond composites, which are excellent candidates for thermal management, has been identified to be determinant of their final properties. In this contribution a new preparative method consisting of electrochemical etching of the metal and spray cleaning, allows microstructural characterization of the interface. This methodology preserves interfacial reaction products and highlights the importance of a proper analysis of both fabrication parameters and sample preparation before any conclusion on reactive selectivity at the interface can be reached.

Keywords: Aluminum/diamond; Metal matrix composites (MMCs); Liquid infiltration; Interface; Electrochemical etching

Most of the relevant properties of composites are determined to a large extent by the characteristics of the host/reinforcement interface. This is certainly the case for composites currently in use, or being developed, for thermal management. Aluminum/diamond composites show outstanding thermal properties [1–7] that may be further improved if the thermal conductance of the interface between the two materials is increased. To this end an unavoidable step is to develop a reliable and reproducible method capable of revealing the interface without affecting its properties. In this case sample preparation is particularly tricky due to the radically different properties of the base materials, diamond and aluminum alloy, and of the most common reaction product (aluminum carbide). In addition, the large hardness that characterizes diamond precludes the use of standard polishing techniques.

The difficulties noted above have forced several research groups to turn their attention to either chemical [4,5] or electrochemical etching [6,7] techniques for preparing samples of aluminum/diamond composites. However, no serious attempt to optimize the experimental conditions has yet been published. Kleiner et al. [6] utilized either electrochemical etching in HNO_3 or chemical etching in HCl, though did not specify the sample-cleaning procedure they used. Ruch et al. [7] electrochemically etched their samples in a 20% aqueous solution of HNO_3 for 20 s, and then cleaned the samples by means of ultrasound in a distilled water bath. Similar techniques have been used in other systems, notably Al/SiC composites [8]. Finding efficient methods for the characterization of materials microstructure requires a deep knowledge of material properties and in most cases a considerable amount of experimental work.

In this context, an interesting point concerns the possible orientation dependence of reactivity in this system. As concluded in Refs. [6,7] reactivity between synthetic diamond single crystals and liquid aluminum seems to be very distinct for the different diamond faces for composites fabricated via gas-pressure infiltration. To rationalize these results the authors argued that while the {111} diamond faces are weakly attacked by liquid aluminum, the two-bonded {100} surface atoms are easily dissolved and participate in the formation of carbides. To which extent this issue is affected by sample preparation is still an open question.

The present work aims to investigate the experimental conditions of a new preparative method to characterize

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reactivity and selectivity at the interface consisting of electrochemical etching of the metal and spray cleaning. This method does not significantly affect the interfacial reaction products. The results indicate that our procedures are probably the most efficient developed up to now, allowing us to conclude that the claimed strong orientation dependence of reactivity is likely a consequence of inadequate sample preparation and/or unsuitable fabrication conditions (mainly infiltration temperature and contact time in the liquid state).

High purity aluminum (99.999 wt.%) was used in this work (supplied by Goodfellow Metals, Cambridge, UK). Preforms were made out of diamond particles of ISD1700 quality, supplied by ILJIN Diamond (Korea); their principal characteristics are reported in Table 1. Particles were packed in graphite crucibles 38 mm long and with a mean diameter of roughly 12 mm, using mixtures of diamond:ethanol in a weight ratio of 15:1. A particle volume fraction of approximately 62% was obtained. Preforms of packed particles were infiltrated with liquid aluminum by means of the gas pressure infiltration technique [9,10]. A solid piece of metal was placed on top of the dried preform and, prior to melting, vacuum was applied in order to facilitate infiltration and reduce porosity. Then, the temperature was raised up to the chosen temperature and argon was introduced into the chamber until a pressure of 5 bars was reached. In this work three infiltration temperatures (760, 800 and 850 °C) were used, namely, 760, 800 and 850 °C, and the contact time t_c (i.e. the time that the composite was kept at the infiltration temperature, and, thus, with aluminum in the liquid state) varied from 0 to 45 min.

Table 1. Characteristic of the diamond particles used in this work.

Diamond (mesh)	$D(90)$	$D(10)$	$D(3,2)$	Span
ISD1700 (40/50)	472	333	392	0.35

D is the average diameter of the particles. The span of the size distribution is defined as $[D(90)-D(10)]/D(50)$, where $D(x)$ is the diameter below which x% of the particles are found. Both D and $D(x)$ are given in μm.

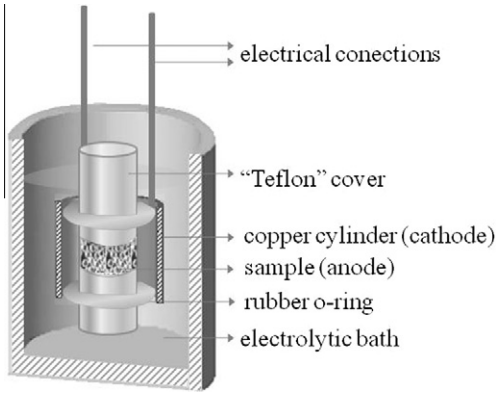

Figure 1. Sketch of the experimental set-up for electrochemical etching of the aluminum/diamond composites investigated in this work.

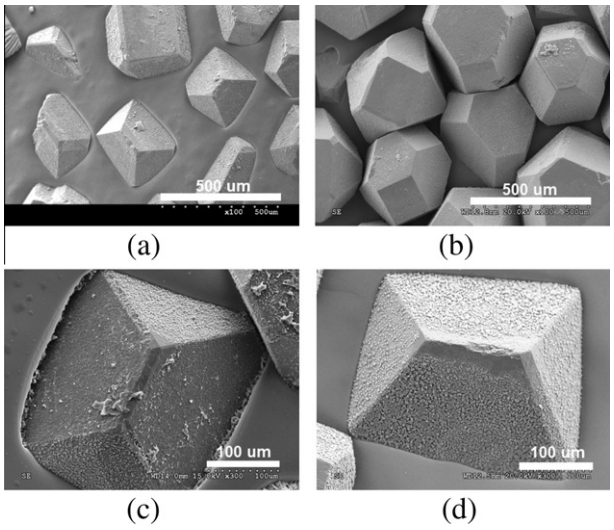

Figure 2. SEM images of diamond particles in an aluminum/diamond composite gas pressure infiltrated at 850 °C for a contact time of 45 min. Electrochemical etching of the composite samples was carried out in a 10% v/v aqueous solution of nitric acid for 1 min (a) 3 min (b) and 2.5 min (c, d). Etched samples were cleaned with acetone either ultrasonically (c) or by spraying (a, b and d).

A sketch of the experimental set-up used for electrochemical etching is depicted in Figure 1. All samples were previously cleaned with ultrasound in an acetone bath for 10 s. The composite (anode) was introduced inside a copper hollow cylinder that acts as the cathode. The radial distance between the sample and the hollow cylinder was fixed at 3 mm using two rubber O-rings (see Fig. 1).

In addition, the upper rubber ring serves for fixing the sample to an electrically conductive (copper) wire. The composite sample was covered with Teflon, allowing only a selected area to be exposed to electrochemical etching. Current density was controlled by a direct current power supply (Blausonic FA-325). Aqueous solutions of nitric acid at concentrations of 5%, 10% and 20% v/v were used as electrolyte, while the current density was fixed at 2 A cm^{-2} (the nominal applied current, in A, was calculated by assuming that the metal matrix represents ~38% of the composite). The relatively large dimensions of the cylindrical samples allow several etching experiments to be carried out on each one by displacing the cathode along its length. After electrochemical etching, samples were cleaned either ultrasonically in an acetone bath for 5 s, or just by spraying acetone at a nominal pressure of 4 bar for 1 min (note that acetone does not dissolve aluminum carbide [11]). Immediately after cleaning, the sample was coated with a thin layer of gold to protect the interface and at the same time to increase image resolution for observation under electron microscopy. Immediate gold coating is essential since aluminum carbide reacts readily with moisture in the air. Direct observation of the interface was carried out, within less than 5 min after coating, by means of scanning electron microscopy (SEM, Hitachi S-3000N).

In discussing the results we should first highlight that the effects of electrochemical etching were similar