Interface Structure in Infiltrated Composites of Aluminum Reinforced with Alumina-Silica Fiber Preforms

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We report here results from an investigation of the interface between fiber and matrix in composites produced by infiltration of alumina fiber preforms containing a silica binder with aluminum. The binder ensures cohesion of the fiber preforms during handling and infiltration. Because it is known that silica is unstable when exposed to liquid aluminum,[1,2,3] the present study was undertaken to assess the extent of reaction of the binder with the matrix when the initial preform temperature is well below that of the liquid metal.

The sample was produced by pressure infiltration, whereby molten metal at 963 K is injected under a constant pressure of 3.6 MPa into a fiber preform contained by a quartz tube and held at 621 K prior to infiltration. The apparatus and casting procedures are described in References 4 and 5. The reinforcement consisted of SAFFIL* fiber preforms with a volume fraction of 24 pct, purchased from Imperial Chemical Industries (Runcorn, United Kingdom). The fiber preform was fabricated by pressing the fibers in the presence of an alkaline solution containing silica. Upon drying and firing of the preform, solid silica deposits on and between the fibers, representing about 5 wt pct of the preform. The fiber itself consists of δ-Al_2O_3 stabilized with an addition of 4 pct SiO_2.

*SAFFIL is a trademark of IC1 Americas, Inc., Wilmington, DE.

Disks 3 mm in diameter and 350 μm thick were cut from the composite sample with a diamond saw and ground mechanically to a thickness of 200 μm. They were subsequently prepared for electron microscopy by dimpling to a thickness of 15 to 20 μm with a diamond paste slurry and milled to perforation with an ion beam accelerated by a potential of 6 kV at an incidence angle of 15 deg. These parameters were then reduced to 4 kV and 11 deg, respectively, to increase the thin area around the initial perforation. Examination was performed with a JEOL 200 CX transmission electron microscope (TEM) at an operating voltage of 200 kV, using a LaB_6 filament and a side-entry double-tilt holder, as well as a VG Microscopes HB5 scanning transmission electron microscope (STEM) operated at 100 kV.

Examination of the sample at low magnification revealed fibers that were essentially intact, surrounded by a fine-grained matrix of aluminum (Figure 1). No second phase was visible within the matrix at this magnification. The fibers are fine-grained polycrystalline, with a grain size of the order of 50 nm, in agreement with previous investigations.[6,7]

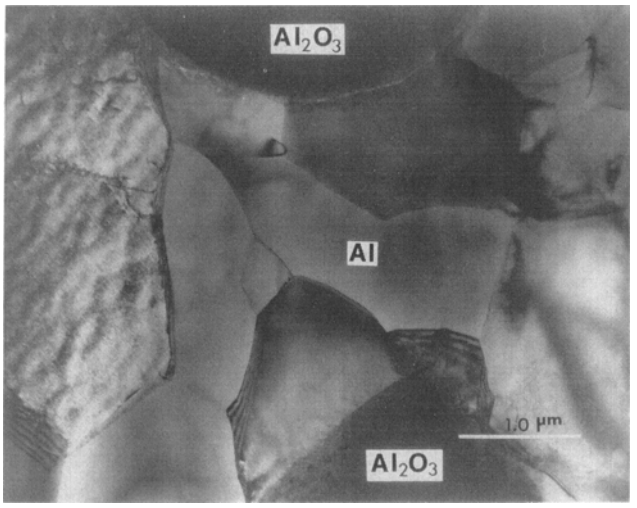

Fig. 1—Low magnification TEM micrograph of the composite sample.

A third phase was found in the form of patches covering somewhat less than half of the fiber/matrix interface (Figure 2) and occasionally bridging two fibers. This phase is devoid of any apparent grain structure and displays a rough, irregular interface in contact with aluminum. A diffraction pattern was collected from a particularly large region of the third phase and showed a two-ring pattern with continuous intensity observed in diffraction mode, distinct from a polycrystalline diffraction pattern, which would contain many more rings of less continuous intensity. This indicates that this third phase was amorphous at the time of observation. The peak intensity diameters of the rings—18.203 and 25.65 mm for a camera constant of 18.4 Å · mm—do not correspond to those of pure quartz in the amorphous

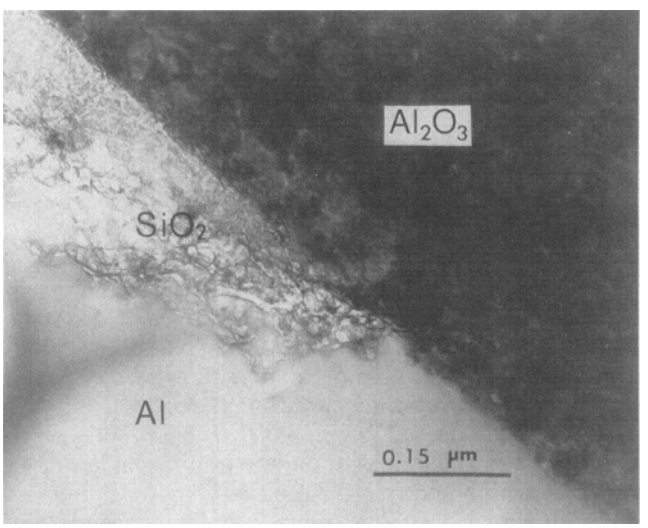

Fig. 2—TEM view of the aluminum matrix, the silica binder, and the alumina fiber.

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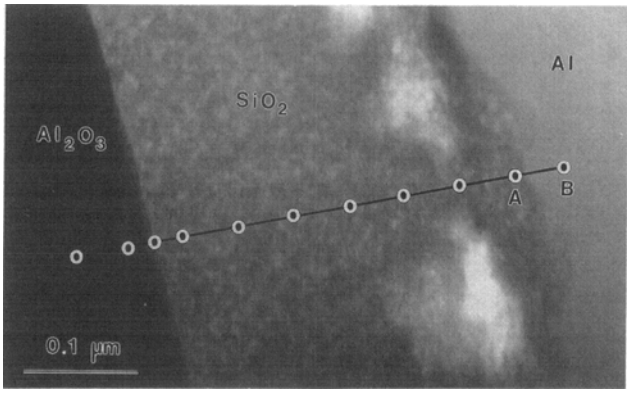
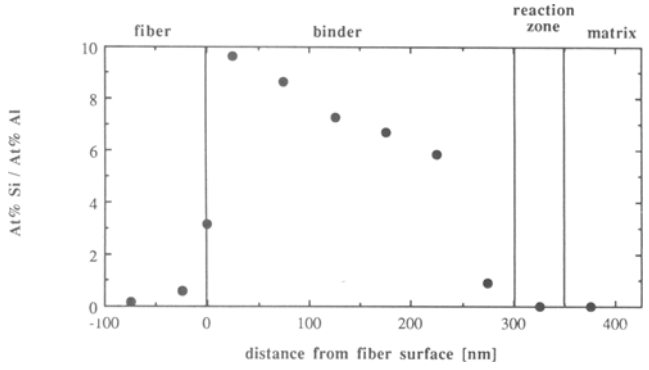
state (obtained in the TEM after beam damage)—21.08 and 31.14 mm for a camera constant of 18.05 Å · mm.[8] Where the binder is absent, the interface is devoid of any reaction products wider than about 5 nm. It is concluded that the fibers themselves do not react significantly with pure aluminum during pressure infiltration with the temperatures and pressure used here, in agreement with previous publications.[7,9]

An elemental dot map of the interfacial phase obtained in the STEM showed that it contains mostly silicon and oxygen and thus constitutes the silica binder, left essentially intact after infiltration. The results of an aluminum and silicon scan through the three regions of Figure 3(a) are presented in Figure 3(b). The observed silicon enrichment in the fiber near the interface is consistent with the as-fabricated fiber structure reported in Reference 9. The binder contains a significant amount of aluminum, which increases as the matrix is approached and explains the discrepancy between the ring diameters measured here and those of pure SiO_2. The outmost region of the binder (point A in Figure 3(a)) is devoid of silicon and shows large amounts of both aluminum and oxygen, while a point in the matrix 50 nm from this layer (point B in Figure 3(a)) exhibits only traces of silicon and oxygen. We thus conclude that the interface between the binder and the aluminum matrix contains a layer of aluminum oxide, presumably resulting from reduction of the silica binder, since no such layer was found at the fiber/matrix interface where the binder is absent (Figure 2). The aluminum-to-oxygen count ratio from this aluminum oxide layer was significantly lower than that collected inside the alumina fiber. Finally, the aluminum concentration profile in the binder suggests that aluminum diffused from the matrix into the binder.



(*a*)



(*b*)

Fig. 3—(*a*) STEM view of the fiber, coating, and matrix, with scanned line of microprobe. (*b*) Ratio of Si/Al (at. pct) along the scanned region of (a).

Thermodynamical data and experimental investigations confirm that silica reacts with aluminum to form alumina and silicon in solution in aluminum.[1,2,3] The low initial fiber temperatures employed in these experiments result in the formation of solid aluminum at the infiltration front.[10] This solid metal may remelt by passage of a remelting front moving from the preform entrance into the composite.[10] Measurements of the metal temperature at the entrance of the mold during similar infiltration experiments with the same apparatus show that superheat in the metal as it reaches the preforms is very small (<5 K) for the initial metal temperature used in casting the sample.[11] It is therefore unlikely that the remelting front has reached the regions investigated in the present study. Fukunga and Goda[12] explained significant reductions in reactivity of amorphous silicon carbide fibers during infiltration with aluminum when the initial fiber temperature was low by the protective action of a layer of solid metal surrounding the fibers. Since silicon carbide fibers feature a silicon oxide layer at their surface and since solid aluminum was most likely present during infiltration in our sample, our observation of limited reaction between silica and aluminum is in agreement with their results.

In conclusion, infiltration by aluminum of a preform kept at a temperature well below that of the metal yields an intimate contact between fibers and matrix. The alumina fiber/aluminum interface exhibits no reaction products. The silica binder initially in the preform is found in patches at the fiber surface and shows limited reaction with the matrix: some aluminum is found in the binder and a layer of aluminum oxide forms at the binder/matrix interface.

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REFERENCES

1. P. Benjamin and C. Weaver: *Proc. R. Soc. London*, 1960, vol. 254A, pp. 177-83.
2. A.E. Standage and M.S. Gani: *J. Am. Ceram. Soc.*, 1967, vol. 50, pp. 101-05.
3. K. Prabriputaloong and M.R. Piggott: *J. Am. Ceram. Soc.*, 1973, vol. 56, pp. 184-85.

4. L.J. Masur, A. Mortensen, J.A. Cornie, and M.C. Flemings: *Metall. Trans. A*, 1989, vol. 20A, pp. 2549-57; 1990, vol. 21A, p. 2287.
5. L.J. Masur, A. Mortensen, J.A. Cornie, and M.C. Flemings: *Proc. 6th Int. Conf. on Composite Materials*, F.L. Matthews, N.C.R. Buskell, J.M. Hodginson, and J. Morton, eds., Elsevier Applied Science, London, 1987, pp. 2.320-2.329.
6. J. Dinwoodie and I. Horsfall: *Proc. 6th Int. Conf. on Composite Materials*, F.L. Matthews, N.C.R. Buskell, J.M. Hodginson, and J. Morton, eds., Elsevier Applied Science, London, 1987, pp. 2.390-2.401.
7. S. Fox and H.M. Flower: data presented at the *E.M.A.G. Conf. of the University of Manchester*, Institute of Science and Technology, Manchester, United Kingdom, 1987.
8. Richard Kontra: Massachusetts Institute of Technology, Cambridge, MA, unpublished research, 1990.
9. G.R. Cappleman, J.F. Watts, and T.W. Clyne: *J. Mater. Sci.*, 1985, vol. 20, pp. 2159-68.
10. A. Mortensen, L.J. Masur, J.A. Cornie, and M.C. Flemings: *Metall. Trans. A*, 1989, vol. 20A, pp. 2535-47.
11. V.J. Michaud and A. Mortensen: Massachusetts Institute of Technology, Cambridge, MA, unpublished research, 1990.
12. H. Fukunaga and K. Goda: *J. Jpn. Soc. Met.*, 1985, vol. 49, pp. 78-83.