Materials Research, Vol. 5, No. 3, 337-342, 2002. © 2002

Aluminium Matrix Composites Reinforced with Co-continuous Interlaced Phases Aluminium-alumina Needles

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Received: September 27, 2001; Revised: July 10, 2002

An Al-5SiO_2 (5 wt% of SiO_2) aluminium matrix fiber composite was produced where the reinforcement consists of fossil silica fibers needles. After being heat-treated at 600 °C, the original fiber morphology was retained but its microstructure changed from solid silica to an interconnected (Al-Si)/Al_2O_3 interlaced structure named co-continuous composite. A technique of powder metallurgy, using commercial aluminium powder and the silica fibers as starting materials, followed by hot extrusion, was used to produce the composite. The co-continuous microstructure was obtained partially or totally on the fibers as a result of the reaction, which occurs during the heat treatment, first by solid diffusion and finally by the liquid Al-Si in local equilibrium, formed with the silicon released by reaction. The internal structure of the fibers was characterized using field emission electron microscope (FEG-SEM) and optical microscopy on polished and fractured samples.

Keywords: *co-continuous structure, silica needles, metal matrix composite*

1. Introduction

In the production of metal matrix composite, one of the subjects of interest when choosing the suitable matrix/reinforcement is the interaction in its interface. In some cases, reactions that occur in the interface between a matrix and its reinforcement have been considered harmful to the final mechanical properties and are usually avoided. In others, the interfacial reactions are intentionally induced when the new layer formed at the interface acts as a strong bonder between the phases[1].

Many works on MMC's published in the last years aimed to obtain a suitable matrix/reinforcement system, that provides minimum and controlled reaction and fine and thermally stable ceramic particulates dispersed uniformly in the metal matrix. These works led to the development of in situ composites, in which the reinforcements are synthesized in a metal matrix by chemical reactions between elements during the composite fabrication. As a result of the process the in situ reinforcement is more thermodynamically stable, the interface between the materials is clean and a strong interfacial bonding is obtained[2].

The mechanisms responsible for in situ formation of ceramic reinforcements in the metal matrix need to be fully understood before commercial use. For the production of metal matrix composites oxide reinforcements have been used, in particles or whiskers morphology, like Al_2O_3, ZrO_2 or ThO_2 in aluminium, magnesium and other metal matrix[3]. Only few works explored the use of silica, SiO_2, as a reinforcement element in non-ferrous matrix. In the aluminium or aluminium alloy matrix, silica had not been used because of the aggressive reactivity between these materials. Preliminary studies[4,5] showed that contact between molten aluminium and silica rich ceramics could destroy completely

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the silica microstructure, due to the reduction reaction $4Al+3SiO_2 \rightarrow 2Al_2O_3+3Si$. Previous works that tried using silica reinforcements in aluminium matrix showed that even at temperatures near 400 °C the silica and aluminium could react and a silica-transformed layer appeared in the original silica surface as a result of the reaction that occurred in the first stage by solid diffusion between the phases[6,7]. Details of the reaction between silica and aluminium and the microstructure of the new layer formed in the silica surface remained unsolved until recent works intentionally induced the reaction by immersing a solid silica body into molten aluminium[8-11]. These works showed that the reaction layer has the form of an interpenetrating metal-ceramic composite, made of an α-Al_2O_3 skeleton filled with an interconnected metal phase that is mostly aluminium with some silicon crystals that remained from the silica reduction and were retained into the alumina channels[12]. Usually this interpenetrating metal-ceramic is known as co-continuous microstructure, in a reference to the continuity of both phases. A well-defined interface is usually observed between the phases Al-Al_2O_3 and between reacted and non-reacted areas (between Al-Al_2O_3 and SiO_2 untransformed). These properties are responsible for elastic-plastic deformations on the composite where the elastic deformation in the ceramic is accommodated by plastic deformation in the metal phase, without significant microcracking[13].

In this study an aluminium matrix composite Al-5SiO_2 was produced using powder aluminium and fossil silica fiber by mixing and hot extruding the mixed components. The bars obtained, after heat treatment in temperatures that did not melt the phases, showed a transformation in the fiber morphology identical to the co-continuous previously discussed. By choosing different heat treatment times, the transformation was induced totally or partially and the microstructure results were compared with the published co-continuous structures obtained by immersion of solid silica bodies into the melted aluminium.

2. Experimental

The mineral silica fiber needles used in this work are cylindrical, tubular, transparent and sharp in both extremities. These fibers are not synthetic, being obtained from natural geological deposits found in abundance in Brazil, named *spongilites*. Their measured diameter and length presented a large normal distribution and were respectively 15.8 µm (standard deviation = 3.2 µm) and 220 µm (s.d. = 125 µm). Its use, in the last few years, has been restricted to the role of filler in the ceramic industry. As the fibers are a result of geological deposits, their composition and crystallography change according to the place where they are taken from in the mine. They are composed predominantly by SiO_2 (> 99.00%), Al_2O_3 (< 0.50%) and Fe_2O_3 (that could reach 1% in some fibers) and show a structure partially amorphous and partially crystalline[14].

The commercially pure aluminium powder was air atomized by *Alcoa* (*PO-101-P*) and presented an equivalent diameter of 9.42 µm (s.d. = 2.42 µm). The energy dispersive spectroscopy (EDS) microanalyses showed 99.70% Al and 0.30% Fe.

The Al-5SiO_2 fiber composite used in this study was prepared by a method previously reported. In this method, the mineral silica fibers (5% in weight) and Al powder were mixed together in a conical blender until the mixture was homogeneous. This aluminium/fibers mixture was conditioned in a commercial aluminium cylinder by cold pressing at approximately 100 MPa, to accommodate the largest amount of material in this recipient and both sides were covered with the same aluminium. After 4.5 h at 450 °C in an electric furnace, the cylinder was hot extruded and its diameter reduced from 100 to 18 mm.

The bar obtained in hot extrusion was heat treated at 600 °C for different periods of time and water-cooled to induce an aggressive interfacial reaction between the matrix and its reinforcements. Heat-treated tests were performed to get a little reaction at the fiber's surface (1 to 2 µm), while the central fiber remained unchanged, or a fully reaction occurred that changed completely the fiber microstructure.

To analyze the reaction induced by heat-treatment, polished samples (grade 400, 600 and 1000 SiC paper, followed by diamond polish in 6, 1 and 0.25 µm and finally polished with colloidal silica) taken from the composite were studied by optical microscopy, SEM (with gold sputtering) and field emission electron microscope FEG-SEM (with carbon sputtering). Additionally, the fracture surface of the composite was analyzed by SEM to show details of the reacted interface. Energy dispersive spectroscopy (EDS) microanalyses were carried out to detect the transformation of silica fiber's composition at the reacted region.

3. Results and Discussion

The extruding process used to obtain the composite was responsible for the alignment of the fibers in the extrusion direction. Sections from the bars in the transversal direction show perfects circular sections and their good distribution in the matrix.

Figure 1, obtained from optical microscopy in a polished sample, shows detail of a silica fiber partially reacted with the aluminium matrix after heat treatment at 600 °C for 1 h. The dark region indicated as A has an untransformed silica composition and in its center a little cylindrical channel (about 1 µm) that always exists in the original silica fiber but with variable width. The region indicated as B shows a typical aspect of the reaction between the fibers and aluminium

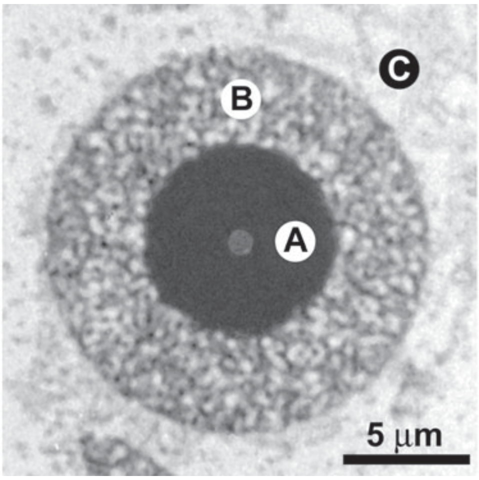

Figure 1. Optical microscopy in a polished sample showing a detail of a fiber partially reacted (B), the center fiber with untransformed silica (A) and the matrix aluminium (C).

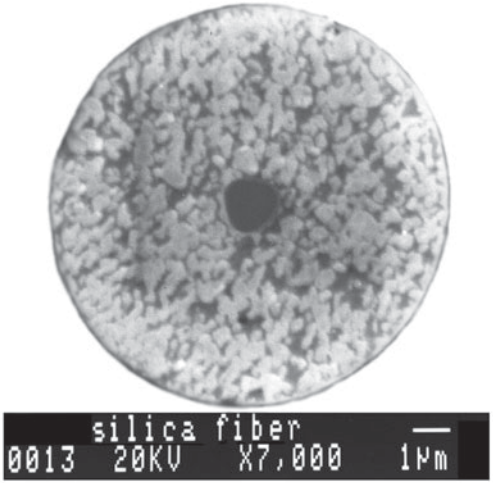

Figure 2. SEM microscopy of transversal polished fiber showing the ceramic phase (white) and the metal phase (dark) with the co-continuous morphology.

(about 4 µm). This region shows the presence of two very refined interlaced phases composed mainly by a metal phase (white phase) and a ceramic phase (dark phase). This reacted region usually has a homogeneous width around the fiber center. The C region indicates the aluminium matrix and some gray phases not sharp in this picture are eutectic Al-Si composition, intermetallic Al-Fe-Si or iron spikes. The picture suggested that there is a good interface between the reacted zone and the matrix and between reacted zone and untransformed silica, but these considerations are restricted by the loss of resolution in the optical magnification.

Figure 2, obtained by SEM, shows a detail of the transversal polished section with a fiber completely reacted. In this picture, the matrix around the fiber was suppressed to clarify the visualization. In the image both phases are clearly seen respectively as a dark phase (metal) and a white phase (ceramic). The appearance of the phases suggest that these microstructures are in agreement with the co-continuous alumina-aluminium micro constituent described in the articles mentioned in the introduction, but in this work their morphology is finer as the temperature used to induce the silica reduction in this work is about a from the ones. The EDS microanalyses at this interconnected micro constituent shows that the SiO_2 original composition was transformed mainly. The picture shows that near the fiber-matrix interface, the new ceramic phase formed is finer with small channels that could connect the metals inside the fiber and the metal from the matrix. The quantity of ceramic fraction could be measured in this picture by image analysis.

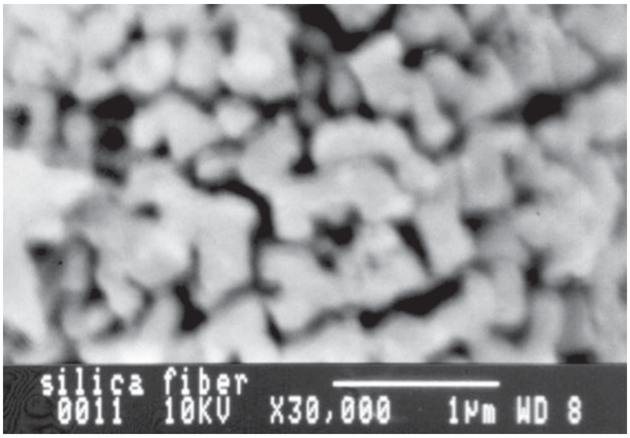

Figure 3. SEM microscopy in the fiber surface after removal of metal phase by HCl immersion, showing arms width of about 0.25 µm.

Figure 3 obtained by SEM in a fiber surface completely reacted after removal of the metal phase by HCl immersion, shows the typical surface morphology with the channels (dark) that were formerly filled with the metal phase. The ceramic phase visualized shows an arms width of about 0.25 µm and homogeneous in the entire fiber surface. Others fibers analyzed show arms approximately between 0.10 to 0.50 µm. This variation suggests that the reaction differ from one fiber to another, probably because of the variations in Fe_2O_3 content previously mentioned.

Figure 4, obtained in FEG-SEM, shows 3 distinct regions in a completely reacted fiber. The black A region in the center fiber is the cylindrical fiber channel previously mentioned. The B and C regions are the reacted fiber microstructure and aluminium matrix respectively. In the B phase, the appearance agrees with co-continuous morphology as in Fig. 2. The gray phase in region B is probably due to the higher Si content than in the matrix composition, that appears in light gray. The same gray phase inside the fiber can be seen at the fiber-matrix interface, suggesting that a liquid Al-Si composition was responsible for infiltrating the channels and that after the reaction is complete some residual Al-Si remains at the interface without diffusing to the matrix.

Figure 5 and 6 were taken from a fractured composite after some reaction was induced. The Fig. 5 shows a fiber strong inserted in the dimple and no pullout occurred. No pull out of the fibers was a usual observation in all fibers analyzed. In this picture the reacted zone can be seen clearly and the 2 phases appear distinct because of the different fracture mechanisms associated. At the left side of this fiber a debonding occurs. Debonding of the fibers occurred in some situations, but the reason for this remains unsolved by this work. Probably some fragile iron-rich phases formed, prevented a good bonding between the metal phases inside and outside the fiber co-continuous zone. Figure 6 is a detail taken from Fig. 5 at the region with good interfacial bonding. The regions A, B and C are respectively the aluminium matrix, the reacted co-continuous region and the silica untransformed. The reacted region shows a good bonding between the aluminium matrix and the co-continuous layer and between the co-continuous layer and the untransformed silica. Because of the different fracture mechanisms at the co-continuous region, some necking of the metal phase can be seen.

Figure 7 gives a schematic model of the changes observed in the fiber microstructure during the heat treatment. The reaction model is divided in 5 steps; according to the time the sample is submitted to heat treatment. In the first stage there are only 2 phases present (silica fiber and pure aluminium). The aluminium starts the reaction with silica according to the equation $3SiO_2 + 4Al \rightarrow 2Al_2O_3 + 3Si$ only by a solid-state diffusion mechanism, and a thin

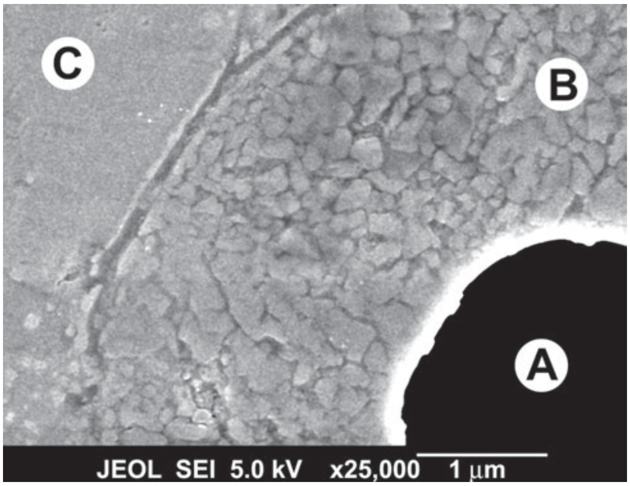

Figure 4. FEG-SEM in transversal polished fiber completely reacted showing the presence of higher Si content at the channels inside the continuous ceramic phase (dark gray at region B) and between matrix (C) and reacted fiber (B).

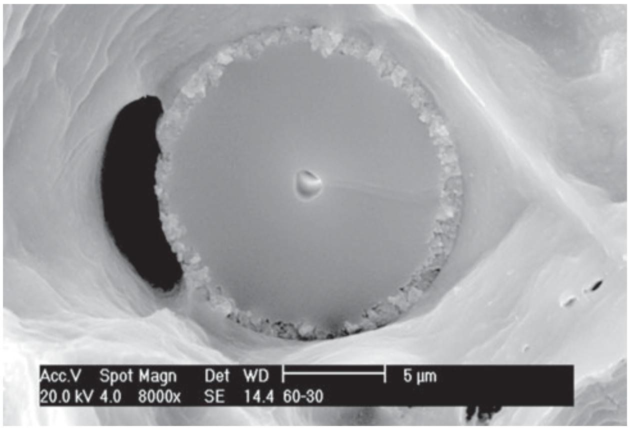

Figure 5. SEM microscopy on fractured composite showing a fiber partially reacted and strong inserted into the dimple. No pullout occurred.

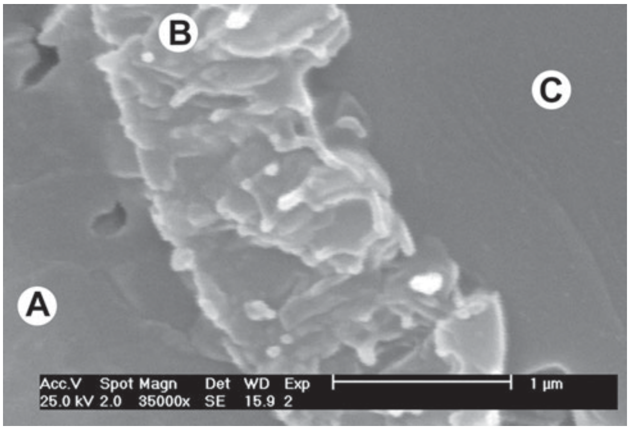

Figure 6. Detail of reaction taken from figure 5 showing aluminium matrix (A), reacted co-continuous region (B) and silica untransformed (C).Some necking of metal phase inside the B region can be see.

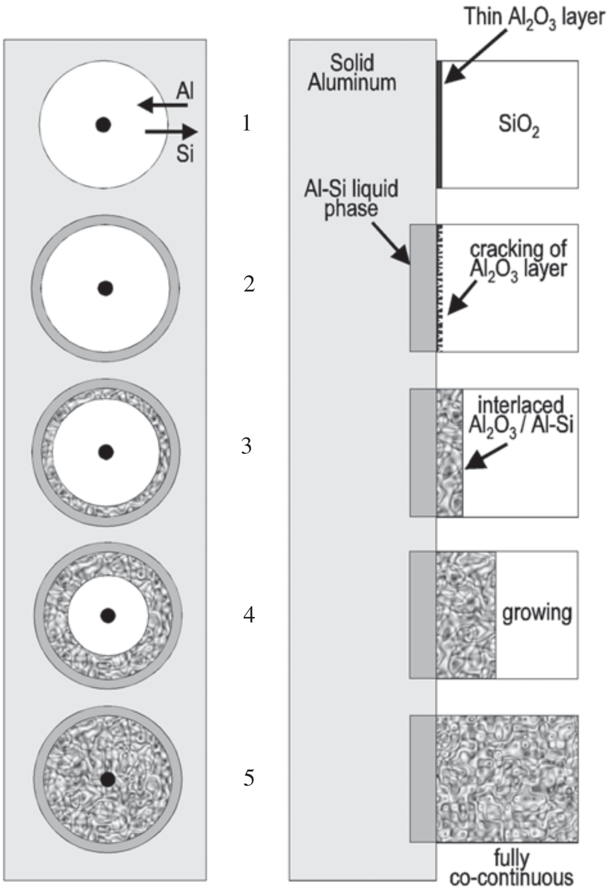

Figure 7. Schematic model of the changes observed in the fiber microstructure during the heat treatment showing the 5 steps that explain the evolution with time of co-continuous morphology.

layer of alumina is formed at the fiber's surface, while some silicon is released to the aluminium matrix direction. At the second stage, the thin alumina layer cracks, as a result of the high tensile stresses induced by the transformation of 3 moles of silica in 2 moles of alumina. At the same time, the silicon released forms, with the aluminium. The liquid Al-Si starts infiltrating the cracks or channels in the alumina and the reaction is accelerated by the liquid dynamic. In stage 3, the thin alumina layer grows and transforms into a co-continuous layer of alumina and Al-Si liquid alloy as the channels create two complex three-dimensional networks. Channels are responsible for the Al transport from the fiber interface to the reaction front and for the silicon transport from the silica into the matrix. Stages 4 and 5 show the growing of co-continuous Al-Al_2O_3 until the fiber is fully transformed and the reaction stops.

4. Conclusions

The possibility of obtaining a co-continuous composite using powder metallurgy as alternative routes has been investigated. The experiments showed that an extruded mixture of fossil silica fibers and commercial powder aluminium heat treated at 600 °C promote a reduction reaction of the silica and the formation of a liquid Al-Si alloy phase, leading to an Al-Si/Al_2O_3 co-continuous structure formed at the original fiber morphology. The well-defined interface present in the composites before the heat treatment was eliminated by this method and the metal occupied the continuous channels created in the new interconnected fiber morphology, promoting a strong bonding between the reinforcements as the matrix metal was connected to the metal infiltrated. The co-continuous obtained showed alumina arms width lower than those obtained by others authors in the immersion of solid silica body in the melted aluminium at higher temperatures and it is affected, probably, by the different Fe_2O_3 content in the fibers.

Acknowledgments

This work was supported by FAPESP under contract n. 00/11374-9. The authors thank *Cepali* and *Alcoa* that supplied the silica fibers and *101-P* powder aluminium respectively. One of the authors (HG) acknowledge partial support through the Conselho de Desenvolvimento Científico e Tecnológico - CNPq.

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