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Oxidation behaviour and effects of oxidation on the strength of SiC-whisker reinforced alumina

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The oxidation behaviour and effect of oxidation on the strength of a SiC-whisker-reinforced-alumina composite material (Al_2O_3-SiC_w) were investigated. The oxidation mechanism of the composite material was determined by thermogravimetric analysis (TGA) and compositional analysis. Changes in the fracture strength and surface morphology were also determined and related to the oxidation mechanism. Weight changes of samples exposed to flowing Ar with various levels of oxygen partial pressure, P_{O_2} at 1400 °C were monitored continuously with a microbalance. Changes in strength were measured after exposure to flowing Ar with different P_{O_2} at 1400 °C for various periods of time. The P_{O_2}-range employed in this experiment was from 5×10^{-7} to 1×10^{-3} MPa. In contrast to the oxidation behaviour of monolithic SiC materials, weight gains were detected for the whole P_{O_2}-range investigated. However, despite the weight gains in the low P_{O_2}-region ($P_{O_2} < 1 \times 10^{-5}$ MPa), significant reductions in strength were observed which were apparently due to the loss of SiC whiskers from the sample surface via the formation of volatile SiO. This SiO gas reacted with the Al_2O_3 matrix to form a non-protective aluminosilicate glass on the surface, resulting in a linear weight gain with time. In the high P_{O_2}-region, typical parabolic weight gains were observed as a result of the formation of an aluminosilicate glass on the surface by a reaction between SiO_2, formed by the oxidation of SiC whiskers, and the matrix alumina. The observed increases in strength of the specimens with exposure are believed to be due to blunting of existing surface flaws with a product oxide.

1. Introduction

Ceramic-matrix composite materials are potentially very useful as high-temperature structural materials because of their better high-temperature mechanical properties compared to monolithic ceramics. Silicon-carbide-whisker-reinforced alumina is one of the most widely studied composite materials, and it is presently being commercially fabricated for wear applications. Even though many researchers have investigated the effects of processing and chemical composition on the strength and toughness [1–4], and the thermal shock behaviour [5], wear resistance [6, 7], creep behaviour [8–11], and brazing behaviour [12] of these composites, the high-temperature environmental stability of these materials has been investigated much less.

An understanding of the environmental stability, especially the oxidation behaviour, of this material is very important for its widespread usage, since many potential applications include long-term exposures to high-temperature oxidative environments. It is well established that the strength and toughness of the Al_2O_3–SiC_w composites increase with the volume fraction of SiC whiskers in the material [4]. Therefore, large amounts of whiskers (about 20 vol %) are generally added to the composite [5–12]. Alumina is stable over a wide range of temperatures and oxygen partial pressures in ambient gas. However, the stability of SiC and other Si-based ceramics, such as Si_3N_4, depends strongly on the exposure temperature and the P_{O_2}-level in the ambient gas [13–18]. A critical point in determining the stability of these materials is the P_{O_2} of the transition from active to passive oxidation. For example, when the partial pressure of oxygen was lower than the transition P_{O_2}, we observed significant reductions in the strength of silicon-based ceramics as a result of material loss through the formation of volatile species such as SiO(g). However, when the oxidant level was higher than the transitional P_{O_2}, increases in strength were generally observed, which were apparently due to the formation of a crack-blunting layer on the surface of the ceramic. In the case of sintered α-SiC, the beneficial layer was identified as SiO_2 [19, 20]; and in the case of a hot-isostatically-pressed Si_3N_4 with 6 wt % Y_2O_3 and 1.5 wt % Al_2O_3 as sintering aids, the layer was identified as either $Y_2Si_2O_7$ or $Y_2Si_2O_7$ and SiO_2

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(depending on the exposure conditions) [21]. Even with small variations in the oxidant level, especially around the transitional P_{O_2}, the room-temperature flexural strengths of both materials varied by more than a factor of two.

The effects of high-temperature exposure on the mechanical strength of SiC-whisker-reinforced alumina, as well as possible oxidation mechanisms, have been reported previously [22–24]. However, most of the exposures were only in strongly oxidizing atmospheres, such as in air in which both of the constituents (Al_2O_3 and SiC) are relatively stable. The oxidation behaviour of the composite as a function of the oxygen partial pressure, especially in environments with low oxygen partial pressures, has not yet been established, and this was the purpose of the research described here. Thermogravimetric analysis (TGA) is generally employed in studying the oxidation mechanism of a material. However, even though it can be a very useful and relatively simple tool for estimating the oxidation mechanism and stability of the material, when used alone it may give misleading results [21]. On the other hand, the high-temperature stability of a material is often estimated by measuring the variation in strength of the material after exposure to air at elevated temperatures for certain periods of times [22]. In this case, the stability of the material in environments other than air, which might be quite different from that in air, is simply overlooked. In this study, the oxidation mechanism and high-temperature stability of an Al_2O_3–SiC_w composite material was investigated by continuously monitoring the sample weight exposed to argon with different levels of P_{O_2}. The variations in room-temperature flexural strength after exposure to argon with various levels of P_{O_2} and after various periods of time were also measured, and they were related to the weight changes observed.

2. Experimental procedure

The SiC-whisker–Al_2O_3 composite materials used in this experiment were hot pressed using a purchased blend of powders consisting of alumina (RCHP-DBM, Al_2O_3 + 0.05 wt % MgO, Reynolds Metals Co., Bauxite, Arkansas, USA), and 20 vol % SiC whiskers (SC-9, Advanced Composite Materials Corp., Greer, South Carolina, USA). The powders were hot pressed in a graphite die for 1 h in a vacuum at 1750 °C under a pressure of 24 MPa. Two billets 63 mm in diameter and 9.5 mm thick were fabricated simultaneously, using a graphite disc as a separator. All the billets had densities higher than 99% of the theoretical density. The samples for weight gain measurements ($0.4 \times 12 \times 12$ mm^3) and bend bars ($2.5 \times 3.7 \times 25$ mm^3) for flexural-strength measurements were cut out from these bulk materials.

The weight changes of samples exposed to argon with various levels of P_{O_2} at 1400 °C were monitored continuously using a TGA system with a microbalance (D-200, Cahn Instruments, Inc., Cerritos, California USA) with a sensitivity of 1.0 μg. A sapphire filament 0.05 cm in diameter (Saphikon, Inc., Milford, New Hampshire, USA) was used in the hot zone to suspend the sample from the balance. This precaution was taken to eliminate (as a source of error) any weight changes that might occur in the wire itself if the metallic support wire extended into the hot zone. The argon gas with different levels of oxygen was introduced from the top of the furnace at a constant speed of 0.9 cm s^{-1} (at standard temperature and pressure, STP) and with a total pressure of 0.1 MPa. The oxygen partial pressure in the ambient gas was controlled by mixing pure argon and argon containing small amounts (100 and 10000 p.p.m (parts per million)) of oxygen, and measured with an oxygen analyser (Model S-3A, Applied Electrochemistry, Inc., Sunnyvale, California, USA). All the gases were dried by passing them through $CaSO_4$ and activated alumina. The humidity level was measured with a hygrometer (Model 3000, Panametric Inc., Waltham, Massachusetts, USA) and it was kept constant during the measurements.

Variations in the strength of the composite after exposure to various P_{O_2}-levels for various periods of time were determined using bend bars cut out from the same batch of billets. The pressed billets were ground and sliced into a flexural-bar shape by attaching them on a stainless-steel holder. The tensile surfaces of the sliced bars (still mounted to the stainless-steel specimen holders) were subsequently polished with diamond pastes down to a grit size of 3 μm. The edges of the tensile surface of each specimen were slightly bevelled on a 6 μm grit diamond lap. After grinding and polishing, the specimens were cleaned in ultrasonically agitated baths of acetone and isopropyl alcohol. Groups of eight specimens were exposed to flowing argon gas with different levels of oxygen for various periods of time at 1400 °C in a resistance-heated, 6.35 cm internal diameter (ID), horizontal alumina tube furnace. The samples, separated by about 0.5 cm, were supported by thin alumina rods to minimize the contact area. The gas flowed parallel to the lengths of the samples, at a constant speed of 0.9 cm s^{-1} (STP) and with a total pressure of 0.1 MPa (slightly above 1 atmosphere). Each sample was weighed before and after exposure to determine the average specific-weight change for the material, and this was compared with the weight change detected by TGA analysis. Four-point-bend flexure tests were performed at room temperature after exposure, using a cross-head speed of 0.008 cm s^{-1}, with inner and outer spans of 6.35 and 19.05 mm, respectively.

The changes in the surface morphology of the samples after exposure were examined by scanning electron microscopy (SEM). The composition of the reaction products formed on the surface was identified by X-ray diffraction (XRD) and energy-dispersive spectroscopy (EDS).

3. Results and discussion

3.1. Oxidation behaviour

Weight changes of the samples exposed to argon with different levels of P_{O_2} at 1400 °C are shown in Fig. 1. When the oxygen partial pressure was very low ($P_{O_2} = 5 \times 10^{-7}$ MPa), a linear weight gain was observed. As P_{O_2} increased to 4×10^{-6} and 1.6×10^{-5} MPa, linear weight gains followed by parabolic weight gains

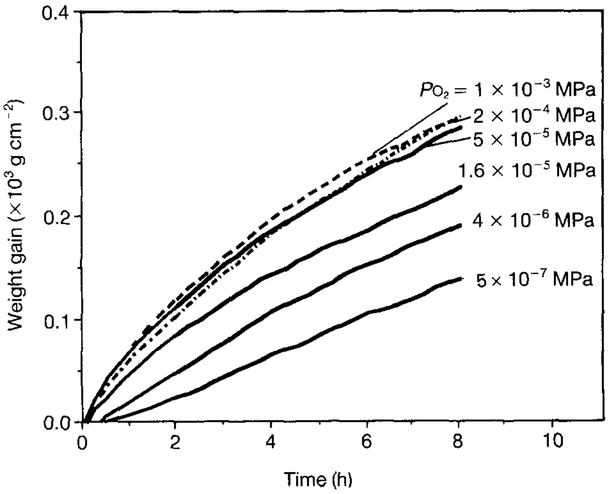

Figure 1 Weight changes of the Al_2O_3–SiC_w samples on exposure at 1400 °C to flowing argon at different levels of P_{O_2}. With $P_{O_2} < 1 \times 10^{-5}$ MPa, linear weight gains with time and an increased rate of weight gain with the P_{O_2} level were observed as a result of the formation of a non-protective aluminosilicate glass on the surface. At $P_{O_2} > 1 \times 10^{-5}$ MPa, parabolic weight gains were observed, and the rate of oxidation was not strongly affected by the P_{O_2} levels.

were observed. When P_{O_2} was higher than 5×10^{-5} MPa, parabolic weight gains were observed with little variation in the weight-gain rate with the P_{O_2}-levels.

It is now well-known that all the Si-based ceramics, such as SiC, Si_3N_4 and the sialons, exhibit two very distinct oxidation behaviours depending on the oxygen partial pressure in the ambient gas [13–18]. When the oxygen partial pressure in the gas is high, that is, in an oxidizing atmosphere, parabolic weight gains are generally detected as a result of the formation of a protective silica layer on the surface. However, when the oxygen partial pressure in the gas is not high enough, material loss occurs via formation of volatile SiO from the sample surface. In this case, the rate of material loss increases with the oxygen partial pressure in the ambient gas until the P_{SiO} at the surface becomes high enough for the formation of SiO_2 on the sample surface.

When a silicon-based ceramic was used as one constituent in a composite material, a similar oxidation behaviour was observed in the high P_{O_2}-range. Parabolic weight gains were detected for the Al_2O_3–SiC composite materials exposed to oxidizing atmospheres (pure O_2 and Ar–1% O_2) at 1375 to 1575 °C [23]. However, the calculated parabolic rate constant, based on the weight-gain data was about three orders of magnitude higher than that of pure SiC. This discrepancy is explained by the fact that the diffusion coefficient of oxygen through aluminosilicate glass, formed by a reaction between silica and matrix alumina, is much higher than that through the pure silica which would form on a monolithic-SiC surface. Therefore, the parabolic weight gains observed in this experiment in the high P_{O_2}-range ($> 5 \times 10^{-5}$ MPa) agree well with the previous results. However, the weight gains observed at a low P_{O_2}-level ($< 1 \times 10^{-5}$ MPa) were rather unexpected because the transition P_{O_2} of SiC from active to passive oxidation (below which weight loss occurs for pure SiC) at 1400 °C is about 1×10^{-5} MPa [17, 19–21]. When the Al_2O_3–SiC_w composite materials were exposed to H_2–H_2O atmospheres at 1400 °C, the oxidation behaviour of the composite was very similar to that of pure SiC; weight loss occurred at a low P_{H_2O} and weight gains at a high P_{H_2O}, with a transition at $P_{H_2O} = 1 \times 10^{-5}$ MPa. In the passive oxidation range ($P_{H_2O} > 1 \times 10^{-5}$ MPa), weight gains occurred as a result of the formation of mullite and aluminosilicate glass as was the case when the composite material was exposed to pure O_2 or Ar–1%O_2. In the active oxidation range ($P_{H_2O} < 1 \times 10^{-5}$ MPa), the SiC whiskers at the surface of the composite material were etched away by the formation of volatile SiO, while the alumina matrix remained intact [24]. Similar results were expected for the composite materials exposed to Ar with P_{O_2}-values lower than 1×10^{-5} MPa. However, contrary to our expectation, weight gains were observed at P_{O_2} lower than 1×10^{-5} MPa, as can be seen in Fig. 1. This phenomenon can be interpreted as a simultaneous vaporization and condensation at the sample surface.

During a study of the active oxidation of silicon in the viscous-flow regime, Hinze and Graham found there are two types of active oxidation depending on the P_{O_2}-level in the ambient gas [16]. When the P_{O_2}-level in flowing gas is sufficiently low, weight loss occurred as proposed by Wagner [25]. However, when the P_{O_2}-level became higher (but still lower than the transition P_{O_2}-value), weight gains were observed by the formation of non-protective, whisker-like SiO_2 on the sample surface. These oxidation products were formed by a reaction between the outward SiO gas and inward O_2 at the gaseous boundary layer. Despite the weight gain detected, this type of oxidation caused severe loss of material because of the consumption of SiO at the boundary layer. A similar phenomenon is believed to have occurred in this experiment. When the $P_{O_2} < 1 \times 10^{-5}$ MPa, SiO gas is formed on the surface of the composite material by the following reaction.

$$SiC_w(s) + O_2(g) \rightarrow SiO(g) + CO(g) \qquad (1)$$

For monolithic SiC, the partial pressure of SiO at the sample surface is not high enough for SiO_2 formation on the surface or in the boundary layer. However, with Al_2O_3 present as the matrix, the P_{SiO} is sufficiently high for formation of an Al–Si–O glass by the following reaction

$$SiO(g) + Al_2O_3(s) + O_2(g) \rightarrow \text{Al–Si–O glass (l)} \qquad (2)$$

During the initial stage of the oxidation, small islands or droplets are believed to form on the surface, as evidenced by the linear weight gain. This type of active oxidation and linear weight gain continues until the liquid phase at the surface covers the whole sample surface. Once the sample surface is covered with the glassy phase, oxygen should diffuse through the reaction-product layer for a further oxidation reaction, resulting in parabolic weight gains. A scanning electron micrograph of the sample surface after exposure to argon with low P_{O_2}(5×10^{-7} MPa) is shown in

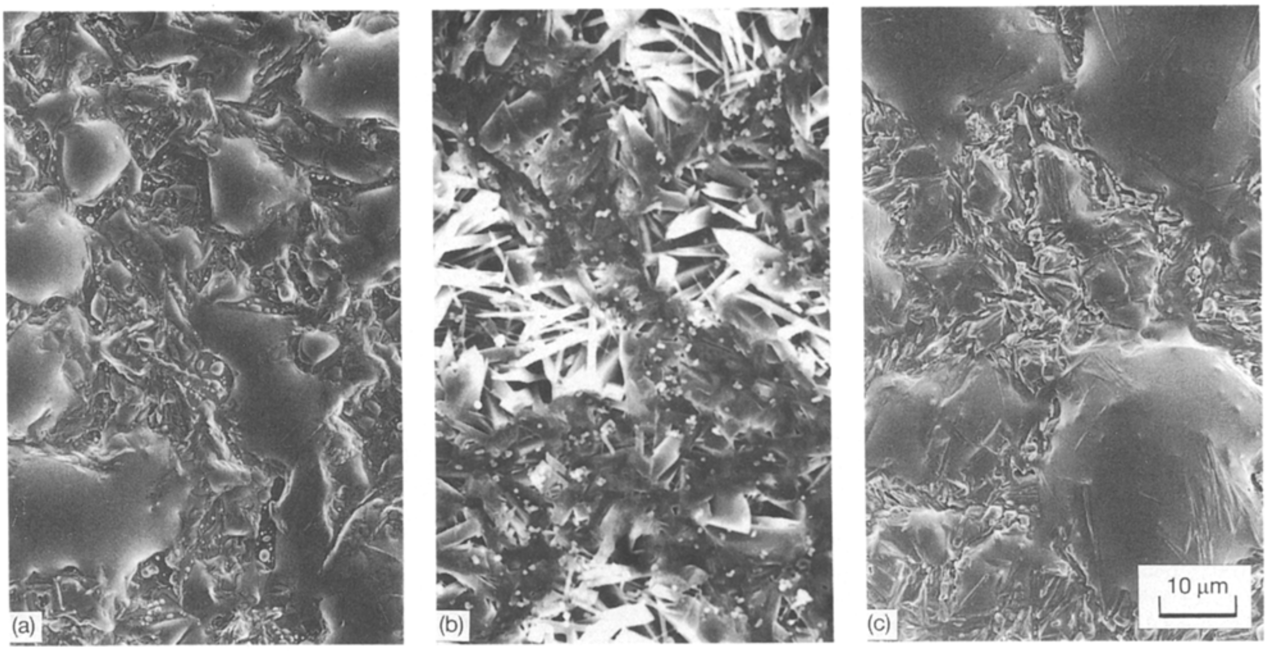

Figure 2 Scanning electron micrographs of the surfaces of Al_2O_3–SiC_w samples after exposure to argon at 1400 °C for 10 h with different levels of P_{O_2} (a) $P_{O_2} = 5 \times 10^{-7}$ MPa, (b) etched with HF solution after exposure to argon with $P_{O_2} = 5 \times 10^{-7}$ MPa, and (c) $P_{O_2} = 1.0 \times 10^{-3}$ MPa.

Fig. 2a. The formation of island-like reaction products on the sample surface is illustrated clearly. Based on their morphology, these reaction products were first thought to be an aluminosilicate glassy phase. However, when etched with a 2.5% HF solution, needle-like grains were revealed, as seen in Fig. 2b. These grains were identified as mullite by X-ray analyses. Comparison of XRD patterns of the samples before and after exposure to argon with $P_{O_2} = 5 \times 10^{-7}$ MPa at 1400 °C, in Fig. 3a and b, respectively, shows clearly the formation of the mullite phase by oxidation. The mullite phase is believed to nucleate and grow from the Al–Si–O liquid during cooling after oxidation. When the samples were exposed to argon with a higher P_{O_2} (still in the active oxidation region for monolithic SiC, that is, $< 1 \times 10^{-5}$ MPa), the surface morphologies as well as the XRD patterns were almost identical. However, when exposed to argon with $P_{O_2} > 1 \times 10^{-5}$ MPa, stronger mullite peaks were detected by XRD analyses, as seen in Fig. 3c. In this case, needle-like mullite embedded in an aluminosilicate glass was also observed by SEM micrographs as seen in Fig. 2c. Even though the composition and morphology of the reaction products of specimens exposed to argon with P_{O_2} higher than 1×10^{-5} MPa are almost identical with those exposed to low P_{O_2}, the routes of product formation are different. As discussed above, for the samples exposed to argon with low P_{O_2}, a glassy Al–Si–O phase is formed via the formation of SiO gas. However, when P_{O_2} is higher than the transitional P_{O_2}, SiO_2 is first formed by the oxidation of SiC and, subsequently, a reaction between this SiO_2 and the matrix Al_2O_3 occurs forming an aluminosilicate glass. In this case, the P_{O_2} in the ambient gas has only slight effect on the oxidation rates, as illustrated in Fig. 1. This implies that even though weight gains were observed for whole P_{O_2}-range, a transition from active to passive oxidation of SiC whiskers has actually occurred in the composite material. This behaviour will affect the strength of the materials, as discussed in next section.

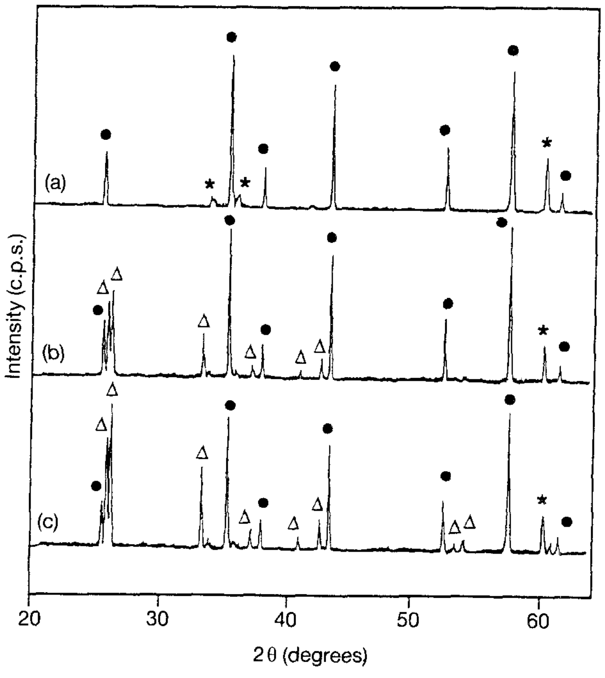

Figure 3 XRD patterns representative of the composite before, and after exposure to argon for 10 h at 1400 °C with various values of P_{O_2} (a) before exposure, (b) 5×10^{-7} MPa, and (c) $P_{O_2} = 1.0 \times 10^{-3}$ MPa. (●) Alumina, (*) silicon carbide, and (Δ) mullite.

3.2. Flexural strength

Changes in the flexural strength of the Al_2O_3–SiC_w composite material after exposure to argon with different levels of O_2 are shown in Fig. 4. Decreases in strength were observed for the samples exposed to argon with a low P_{O_2}. However, when the P_{O_2} in the ambient gas was higher than 1.5×10^{-4} MPa, increases in strength were observed. This trend was also

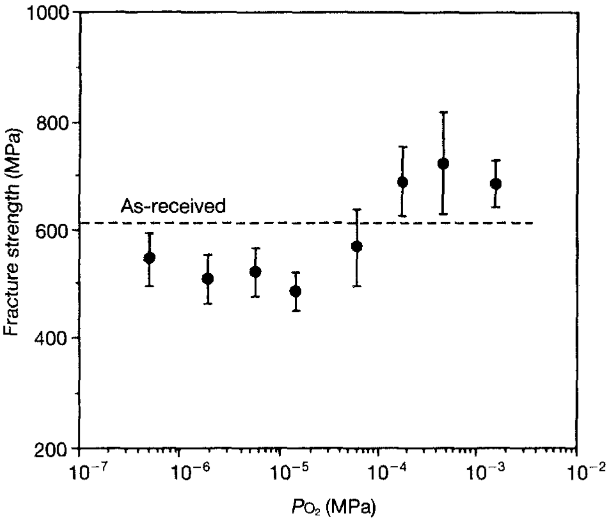

Figure 4 The average room-temperature fracture strengths of the Al_2O_3–SiC_w composite materials exposed for 10 h at 1400 °C to flowing argon with various levels of P_{O_2}. The error bars represent plus or minus one standard deviation.

observed in other silicon-containing monolithic materials, such as SiC and Si_3N_4[14, 19–22, 24]. However, for those monolithic materials, the strength variations were proportional to the weight change of the samples; reductions in strength occurred with weight loss in the samples, while increases in strength occurred with weight gains (at least in the early stages of oxidation). In the present case, reductions in strength were detected in the samples with weight gain. The reductions in strength in this P_{O_2} range ($P_{O_2} \leqslant 1.5 \times 10^{-5}$ MPa) are apparently due to the loss of the SiC whiskers used as a toughening/strengthening agent. As discussed above, in this P_{O_2}-range the SiC whiskers in the composite react with O_2 to form SiO gas, which in turn reacts with the matrix Al_2O_3 forming an Al–Si–O liquid phase. Therefore, despite the weight gains observed, significant amounts of SiC whiskers at the surface of the composite were lost, leaving the spaces that had been occupied by the SiC whiskers as surface defects. The reductions in strength became more severe as P_{O_2} increased as a result of the increased loss rate of SiC whiskers from the surface. A minimum in strength was detected at $P_{O_2} = 1.5 \times 10^{-5}$ MPa, which is very close to the transition P_{O_2} from active to passive oxidation of monolithic SiC. These results support the premise that even though weight gains were observed for the whole P_{O_2} range, transition from active to passive oxidation actually occurred.

When the P_{O_2} was increased to values higher than 1 $\times 10^{-4}$ MPa, increases in strength were observed, as seen in Fig. 4. Increases in strength are apparently due to the formation of continuous protective layers on the surface. When the composite material is exposed to the high P_{O_2}-range, SiC whiskers in the composite are oxidized first to form SiO_2 which subsequently reacts with the matrix alumina to form an Al–Si–O liquid. Therefore, in contrast to the oxidation behaviour in the low P_{O_2}-range, reaction products are likely to be formed around the whiskers without forming voids near the surface. The reaction-product layer blunts the flaws at the surface, resulting in an increase in the strength of the material. Similar trends have been observed previously for other silicon-containing monolithic and composite materials exposed to high P_{O_2} or P_{H_2O} atmospheres [19–22, 24].

The influence of the exposure time on the strength of samples exposed to argon with two different P_{O_2} levels at 1400 °C are shown in Fig. 5. When exposed to argon with a low P_{O_2} (1.5×10^{-5} MPa), the strength decreased with the exposure time up to 10 h and remained constant after further exposure. These strength variations reflect well the oxidation mechanism proposed above; that is, non-protective aluminosilicate islands are formed on the surface after the consumption of SiC whiskers, until the product layer covers the sample surface completely. As SiC whiskers are lost by the reaction with O_2, the size of critical defects near the sample surface increases with the reaction time, leading to a decrease in the strength of the specimens. However, once the whole surface is covered with an aluminosilicate layer, formation of SiO gas is suppressed, so that the size of critical defects and, accordingly, the strength of the specimens remains constant.

When the P_{O_2} in the ambient gas was 5×10^{-4} MPa, which is higher than the transitional P_{O_2} of SiC at 1400 °C, increases in strength were observed for samples exposed for times up to 20 h. However, as seen in Fig. 5, when the materials were exposed for more than 20 h, the strength of the samples after exposure was about the same as the strength before exposure. This phenomenon of a rapid increase followed by a gradual decrease in strength with exposure time is frequently observed in other silicon-based materials [14, 20–22]. It is generally accepted that the increase in strength is due to the blunting of existing surface flaws, as mentioned above, while the decrease in strength is due to the formation of new defects such as bubbles or cracks at the surface. When the reaction layer becomes too thick, product gases such as SiO or CO cannot escape rapidly enough through the product layer; this causes the formation of bubbles in the layer. These

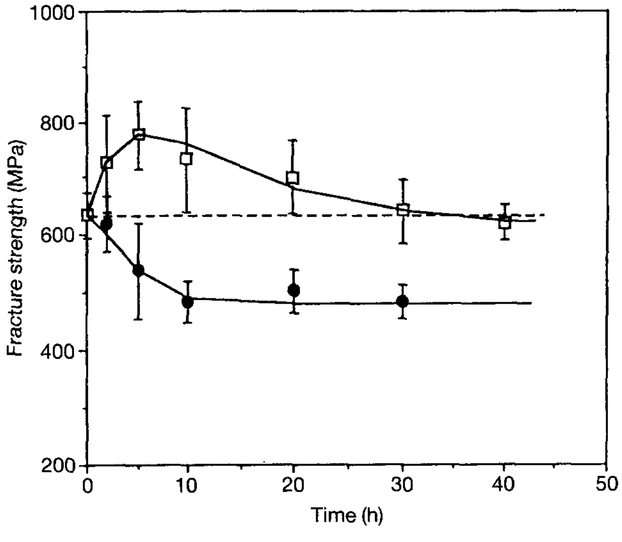

Figure 5 The effects of exposure time on the strength of the composite material exposed at 1400 °C to Ar at two different levels of P_{O_2}: (□) 5×10^{-4} MPa, and (●) 1.5×10^{-5} MPa.

bubbles might act as critical defects when specimens are under tensile stress. Another source of new defects is the difference in the thermal-expansion coefficients between the reaction products and the substrate. When the thermal-expansion coefficient of the reaction products on the surface is larger than that of the substrate, residual tensile stress is developed in the layer during cooling. This residual stress may generate cracks in the product layer (which sometimes penetrate into the substrate, leading to a remarkable reduction in strength [26, 27]). Even though not generating cracks, the crack-blunting effect due to the formation of this layer is negated by this residual stress. In the present case, the thermal-expansion coefficient of the aluminosilicate glass formed on the surface by the oxidation process is presumed to be higher than that of the Al_2O_3–SiC_w composite material. Therefore, on oxidation of the material, two competing processes are operating simultaneously: crack blunting, which leads to an increase in the strength; and formation of bubbles or the development of a residual tensile stress, which leads to a reduction in the strength of the material. At the initial stage of oxidation, an increase in strength was observed because the crack-blunting effects are dominant. As the product layer becomes thicker with exposure time, the probability of bubble formation in the layer increases, and at the same time a higher residual tensile stress is developed in the surface, leading to a reduction in strength of the material. The observed variations in the strengths of the samples, seen in Fig. 5, reflect these compositional and microstructural evolutions of Al_2O_3–SiC_w composite materials exposed to argon with P_{O_2}-levels higher than the active to passive transition value.

4. Conclusion

The oxidation behaviour and high-temperature stability of Al_2O_3–SiC_w was determined by monitoring the changes in weight and strength of the samples exposed to flowing argon containing different levels of oxygen. These characteristics were strongly affected by the P_{O_2}-level in the ambient gas and by the exposure time. For the whole P_{O_2}-region investigated (from 5×10^{-7} to 1×10^{-3} MPa), weight gains were observed. When the oxygen partial pressure was lower than the transition P_{O_2}, from active to passive oxidation of monolithic SiC (1×10^{-5} MPa), linear weight gains were observed and the rate of weight gains increased with the P_{O_2}-level in the ambient gas. However, in spite of the weight gains in this region, reductions in strength were observed. These reductions in strength became larger with the exposure time up to 10 h and remained constant for further exposure. These reductions in strength are due to the loss of SiC whiskers from the sample surface via the formation of volatile SiO. This SiO gas reacted with the matrix Al_2O_3 to form a non-protective aluminosilicate glass on the surface, resulting in linear weight gains with time. As the reaction time increased, the amount of the liquid phase increased to cover the sample surface completely, causing parabolic weight gains and no further reduction in strength. When the P_{O_2} was higher than the transition P_{O_2} ($> 1 \times 10^{-5}$ MPa), typical parabolic weight gains were observed, and the rate of oxidation was not much affected by the P_{O_2}-levels. In this P_{O_2}-range, the variations in strength after oxidation were also very similar to those observed for monolithic SiC, that is, a rapid increase followed by a gradual decrease in strength with exposure time. The reaction products formed on the surface were identified as a mixture of aluminosilicate glass and mullite.

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