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Corrosion of a unidirectionally solidified Al_2O_3/YAG eutectic composite in a combustion environment

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Abstract

Owing to their remarkable higher creep resistance, some oxides eutectic composites those fabricated by unidirectional solidification are prime candidates for structural components used in a severe corrosive environment at high temperatures. In this paper, the possibility of an Al_2O_3/YAG eutectic composite for high-temperature application, where the materials would be exposed to combustion gases, was investigated. The Al_2O_3/YAG eutectic composite was stable at 1700 °C in an atmosphere of oxygen/water vapor (O_2/H_2O), showing only slight changes in microstructure, volume and flexural strength after an exposure for 200 h. Thus, Al_2O_3/YAG eutectic composite is among the most promising ceramics for structural applications at high temperatures.

Keywords: Composites; Corrosion; Engine components; Microstructure-final

1. Introduction

In order to improve the reliability of ceramics, some new material systems have been developed, which have the ability to produce microstructures in situ with unique micromorphology that allow toughening mechanisms to take place. These self-reinforced ceramics can be formed by (i) growth from the liquid phase (e.g., $Si_3N_4Al_2O_3$ and mullite);[1–3] (ii) by decomposition of single solid solution (e.g., AlN−SiC);[4] and by (iii) simultaneous crystallization of more than one solid from a eutectic composition liquid (e.g., directionally solidified oxide eutectic composites those we call as melt-growth-composites, MGCs).[5] In all cases, the initial composition forms a liquid or solid solution with a different morphology. The preferred crack propagation along interface boundaries makes them more flaw-tolerant than monoliths. The availability of common raw materials, lower production cost than that of the fiber composites, and good controllability of phase morphology are also their advantages.

The major attractive feature of MGCs lies in improvement of the creep property at high temperatures. Polycrystal ceramics are usually composed of many grains with different orientations. A grain boundary has excess energy, therefore, grain growth and grain boundary diffusion creep will easily progress at high temperatures. In the extreme case, superplasticity occurs due to grain boundary sliding if the ceramics contain a widely distributed non-crystalline grain boundary film. In contract, MGCs have neither grain boundaries nor colonies as can be seen in Fig. 1.[6] This unique microstructure is grown by the unidirectional solidification from ceramic melts with a eutectic composition which minimizes the free energy at the interface, resulting in single crystal phases entangled with each other.

In addition, locally distributed thermal residual stresses develop in the eutectic composite upon cooling from the solidification processing temperature as a result of the mismatch in thermal expansion coefficients between both phases (in the case of binary eutectic composite). The compressive residual stresses if they were formed in the more brittle phase of the eutectic composite will contribute actively to increase its strength. Because, failure is likely to start in the weaker

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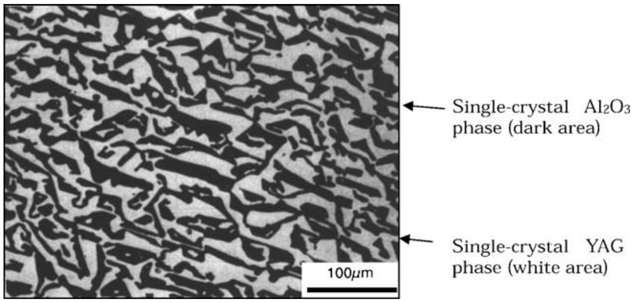

Fig. 1. Microstructure of a unidirectionally solidified Al_2O_3/YAG eutectic composite (melt growth composite, MGC).

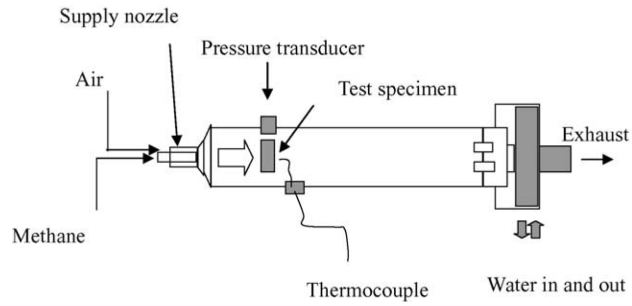

Fig. 2. Schematic drawing of a burner rig test.

phase but this requires overcome the compressive residual stresses.

On the other hands, oxide ceramics have been widely used owing to their environmental stability. However, the chemical property data of the oxide eutectic composites being formed the anisotropic residual thermostresses locally, needed to predict criteria for corrosion, are less available in the literature. Environmental resistivity, the ability to withstand high-operating temperatures, atmospheres and/or attack by corrosive melts, is an important property of materials for practical use. In this study, the stability of the $Al_2O_3/Y_3Al_5O_{12}$ (YAG) binary MGC in water vapor at 1700 °C was investigated. The stability in combustion field at 1500 °C was also investigated.

2. Experimental procedure

The MGC material (Al_2O_3/YAG eutectic composite) used in this work was supplied from Ube Industries Ltd., Japan. It was fabricated by melting the mixture powder of Al_2O_3/Y_2O_3 = 0.82/0.18 in mole ratio, and solidifying directionally in a molybdenum crucible using a Brigman technique (the growth rate: 5 mm h^{-1}) with approximately 40 mm diameter and 70 mm long. The samples for the corrosion test were cut into rectangular solid of bars (3 mm × 4 mm × 40 mm) and plates (10 mm × 10 mm × 1 mm), and all of the surface were polished to a 6-μm finish, and ultrasonically cleaned in pure analytical grade ethanol. Typical microstructure of the Al_2O_3/YAG (0.52/0.48 in volume ratio) eutectic composite shows the two single crystal phases complexly interpenetrated without colony structures and fibers as in conventional directionally solidified materials (as in Fig. 1).

Corrosion tests in wet-O_2 gas were conducted up to 200 h at 1700 °C in a horizontal tube furnace equipped with an Al_2O_3 tube of 99.8% purity. The bar samples were placed on high purity Al_2O_3 setters supporting the pinpoints near both edges of the sample in order to expose all of the sample surface. The sample temperature was monitored with a Pt–20% Rh thermocouple placed within 10 mm apart from the sample. The heating rate from room temperature to the test temperature of 1700 °C was 250 °C h^{-1}. The Al_2O_3 tube and the setter had been preheated at 1700 °C for 50 h passing the wet-O_2 gas before the use for the corrosion tests.

The corrosive atmosphere was H_2O: 5.0×10^4 Pa and O_2: 5.0×10^4 Pa under atmospheric pressure. The gas flow rate was 3.3×10^{-6} m^3 s^{-1} in total. Water vapor was introduced into the oxygen stream using a saturator where oxygen was bubbling through deionized water held at 82 °C.

In general, experiments of an alumina sample exposure to the wet atmospheres may be limited by mullite formation at the high end of the temperature range below 1500 °C.[8] Volatile Si–O–H species are often carried from the fused quartz tube to the alumina sample surface where mullite is nucleated. In this study, therefore, the components of the furnace where hot water vapor passing was made with high-purity alumina only, so that the Si–O–H contamination could be prevented. Mullite was not observed at any exposure temperatures up to upper limit temperature of the furnace (1700 °C) in this study.

After the corrosion experiments, the weight and volume changes were measured. The three-point bending test of the corroded samples was made at a cross-head speed of 0.5 mm min^{-1}. Most of the data were averaged ones of three to four values. scanning electron microscopic (SEM) and electron microprobe Analyses (EPMA) were made to observe the microstructures and to study their chemistry. X-ray diffraction (XRD) was also used to determine the phase composition. Commercially available sintered polycrystalline Al_2O_3 (SSA-S; Nikkato Co., Japan) was also tested for comparison.

The burner rig test was conducted for examination of the effect of combustion gases including CO, CO_2, H_2 and H_2O, on the sample recession. Fig. 2 shows a schematic of the high-pressure burner rig configuration. This burner rig was originally developed for an investigation of CH_4 combustion at high-pressures. The sequence of CH_4 combustion such as ignition of fuel gas, pressure controlling during combustion, extinguishment and forced cooling by air, is accomplished using a computer-tip-based digital data acquisition system. The ceramic samples exposed by this burner rig often resulted in crack because of its poor thermal shock resistivity, therefore, the slight change at the sample surface caused by the attack of the combustion products (i.e., radicals) was mainly investigated.

Air and CH_4 are supplied from gas cylinders to this burner rig. CH_4 is supplied through the inner nozzle of a coaxial