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Microstructure and mechanical properties of a directionally solidified $Al_2O_3/Y_3Al_5O_{12}/ZrO_2$ hypoeutectic in situ composite

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ABSTRACT

Directionally solidified ternary $Al_2O_3/Y_3Al_5O_{12}$(YAG)/ZrO_2 hypoeutectic rod composites were successfully fabricated by the laser zone remelting technique. The microstructure and mechanical properties of the composite were investigated. The microstructure presented a complex three-dimensional network structure consisting of fine Al_2O_3 (41 vol.%) and YAG (49 vol.%) phases, with smaller ZrO_2 (10 vol.%) phases partially distributed at the Al_2O_3/YAG interfaces. The irregular growth behavior in the hypoeutectic was revealed. The hardness and fracture toughness at ambient temperature were measured to be 17.3 GPa and 5.2 MPa $m^{1/2}$, respectively. The toughness enhancement in comparison with previous binary Al_2O_3/YAG composites was mainly attributed to the refined microstructure, and crack deflection, branching and bridging. Moreover, the residual stresses, generated by different thermal expansion coefficients of the component phases, also importantly contributed to the improved toughness. Correlations between the addition of the third component ZrO_2 and the microstructure and properties were discussed as well.

1. Introduction

In recent years, a great demand for higher temperature capabilities in aerospace and power systems has continued to motivate researches on advanced high temperature structural materials, which requires both superior mechanical strength and oxidation resistance at temperatures above 1923 K [1–3]. Because of the unique attractiveness in outstanding oxidation resistance, high stability of the microstructure and excellent mechanical properties retention (almost constant up to temperatures close to the melting point) in oxidizing environment, directionally solidified oxide eutectic in situ composites stand out as promising candidate materials for structural applications at elevated temperatures [4]. Moreover, the lower densities of oxide eutectics and their higher use temperatures, as well as a reduced or no need for cooling medium, provide for improved thermal efficiency and high temperature performance compared to conventional Ni-based superalloys. As a result, these advantages prompt an extensive search and development of new oxide eutectic in situ composites. Up to now, several directionally solidified oxide–oxide eutectic composites, such as Al_2O_3/YAG [5–9], $Al_2O_3/GdAlO_3$ [10], $Al_2O_3/Er_3Al_5O_{12}$ [11], Al_2O_3/ZrO_2 (Y_2O_3) [12] and $Co_{1-x}Ni_x/ZrO_2$ (CaO) [13], have been widely developed. Among these, directionally solidified Al_2O_3/YAG eutectic in situ composite is currently regarded as one of the most promising candidates for future generations of ultra-high-temperature structural materials due to its high melting point (T_m = 2100 K), substantially low density (4.1 g/cm^3), excellent high-temperature strength (up to 2073 K), microstructural stability, creep resistance, corrosion resistance and oxidation resistance [5–9]. Additionally, the Al_2O_3/YAG eutectic composite can also be used as fiber reinforcements with excellent properties in metal-matrix composites [14,15]. Nevertheless, it is well known that most of ceramic materials generally suffer from a catastrophic failure due to their low toughness and poor ductility. Likewise, it has no exception for Al_2O_3/YAG eutectic composites. The fracture toughness of Al_2O_3/YAG at ambient temperature is very low (~2 MPa $m^{1/2}$) [7]. Therefore, their application range in practice is extremely restricted.

It is recently expected that the development of oxide eutectic composites, which well combines flaw tolerance, strength and toughness, is one of the feasible approaches to efficiently achieve a desirable balance between room temperature ductility and high-temperature strength [4]. In this respect, it is essential for real applications in improving the strength and toughness of directionally solidified eutectics. At present, there are two prime approaches to actualize this objective. One is to refine the microstructure and reduce the flaw size through microstructural control mechanism such as using rapid solidification. For instance, a better toughness (3.6 MPa $m^{1/2}$) has been reported in the Al_2O_3/YAG eutectic composite at high growth rates due to the fine and homogenous microstructure in the submicron range [16]. Another important way for improving the toughness is the incorporation of the third component such as ZrO_2 into the binary eutectics to form ternary composites. The brittleness of the binary Al_2O_3/YAG eutectic is believed to be mainly caused by the strong interface bonding

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between the two constituent phases besides their inherent brittle nature [4]. The interface could be weakened by the third phase. The crack paths are possible to be altered to promote crack deflection or microcracking, and consequently, obtaining significant toughening. A much more improved toughness (~8–9.0 MPa $m^{1/2}$) has been obtained in the Al_2O_3/YAG/ZrO_2 ternary eutectic composite [17]. Great attention has thus been addressed towards the ternary eutectic composites recently [18–20]. However, in the case of the ternary Al_2O_3/YAG/ZrO_2 eutectic composite, it has been noticed that the eutectic solidification range is narrow, and the composition volatilization at high temperature is generally produced [19]. So, it is more required to investigate the ternary composite off to the eutectic composition. On the contrary, it is limited report till date. Furthermore, the study also favors further understanding of the complex solidification behavior of the ternary oxide eutectic and to optimize the mechanical properties. In fact, the eutectic structure can not only be obtained in the eutectic composition but also at the off-eutectic composition if appropriate growth conditions are carefully selected [21].

Laser zone remelting is a recently developed technique used in producing directionally solidified oxide eutectic composites [16,22,23]. Compared with conventional directional solidification methods, the exceptional advantages of this technique rely on the nonexistence of crucible (without contamination), high melting temperature and large thermal temperature gradient (~10^6 K/m), and consequently, high growth rates can be obtained. In addition, the microstructure can be controlled in a wide range of the growth rates during laser zone remelting, achieving an optimization in mechanical properties. In the present study, the directionally solidified ternary Al_2O_3/YAG/ZrO_2 hypoeutectic composite were fabricated by using laser zone remelting. The emphasis was on examining the microstructure, mechanical properties and toughening mechanisms.

2. Experimental

2.1. Sample preparation

Specimens of the directionally solidified Al_2O_3/YAG/ZrO_2 hypoeutectic composite were prepared using a mixture of high-purity (>4 N) nano-powder of Al_2O_3, Y_2O_3 and ZrO_2. The selected mole ratio of the hypoeutectic composition was $Al_2O_3/Y_2O_3/ZrO_2$ = 71/17/12 [24]. Precursor rods (Φ 7 mm × 60 mm) were isostatically pressed at room temperature and then sintered at 1773 K for 2 h in order to improve handling strength. Directional solidification was performed in a vacuum environment using the laser zone remelting technique with a 5 kW CO_2 laser as a radiation source, at a laser scanning rate range from 10 to 2000 μm/s. Further detailed processing procedure has been described elsewhere [16]. The final rods of the hypoeutectic composite with about 6 mm in diameter could be obtained.

2.2. Microstructure characterization

Microstructural observations and chemical analyses of the composite were determined by scanning electron microscopy (SEM, JSM-5800), energy disperse spectroscopy (EDS, Link-Isis) and X-ray diffraction (XRD, Rigakumsg-158) techniques. Quantitative calculation of the phase volume fraction was performed by digital image analysis software of SISC IAS V8.0.

2.3. Mechanical tests

Hardness and fracture toughness were measured by the Vickers indentation test technique. The indentations were made using 9.8 N loads for 15 s, and at least ten valid microindentations were conducted in each sample. The hardness and fracture toughness are calculated according to the following equations proposed by Niihara [25] for Palmqvist cracks:

$$H = k_1 P/a^2 \quad (1)$$

$$K_{IC} = k_2 (E/H)^{2/5} (p/al^{1/2}) \quad (2)$$

where H is the Vickers hardness, K_{IC} the fracture toughness, E the Young's Modulus, P the indentation load, a half the indentation diagonal, l the crack length, and k_1 and k_2 the dimensionless constants determined experimentally, respectively. The SEM was used to observe the crack propagations and to ascertain the toughening mechanism.

3. Results and discussion

3.1. Characteristic microstructure

Fig. 1a and b show the typical microstructures of the transverse sections of the laser remelted ternary Al_2O_3/YAG/ZrO_2 hypoeutectic composite grown at the scanning rate of 10 μm/s. The sintered composite with the same composition is shown in Fig. 1c. For comparison, the laser remelted Al_2O_3/YAG/ZrO_2 ternary eutectic and Al_2O_3/YAG binary eutectic composites grown at the same growth conditions are shown in Fig. 1d and f, respectively. The XRD patterns of the hypoeutectic composite are shown in Fig. 1e. Combining the EDS analysis, it is clearly indicated that the Al_2O_3/YAG/ZrO_2 hypoeutectic rods show only three distributed phases: Al_2O_3 phase (dark zone), YAG phase (grey zone) and cubic Y_2O_3 stabilized ZrO_2 phase (white zone). No other crystalline phases are formed or amorphous signal is detected. The microstructure of the composite presents a typical off-eutectic microstructure with three-dimensional network structure, consisting of a large primary crystal (Al_2O_3/YAG binary eutectic) and a fine Al_2O_3/YAG/ZrO_2 ternary eutectic. The volume fractions of three component phases, as measured by the digital SEM images, are 41%Al_2O_3, 49%YAG and 10%ZrO_2, respectively, which is well consistent with that predicted for the hypoeutectic composition through the phase diagram [24]. Comparing the volume fractions with the ternary eutectics (40%Al_2O_3, 43%YAG and 17%ZrO_2), the result shows that it is possible to change the relative volume fractions of the eutectic phases by going to off-eutectic compositions. In the hypoeutectic composite, the Al_2O_3 and YAG phases are interconnected each other, and the ZrO_2 phases are partially distributed at the Al_2O_3/YAG interfaces or at the edge of the YAG phases. The sizes of ZrO_2 phases are much smaller than Al_2O_3 and YAG phases. Furthermore, it is noted that the hypoeutectic microstructure is similar to a cellular structure appeared in the quasi-eutectic Al_2O_3/YAG/ZrO_2 ternary composites [26] and the Al_2O_3–ZrO_2 (Y_2O_3) binary eutectics [27] reported in previous literature. The formation is believed to be the consequence of the addition of the third element (off-eutectic composition) and interface instability in the solidification front during laser zone remelting [22,26]. Moreover, there are no pores or grain boundaries found in the Al_2O_3/YAG/ZrO_2 hypoeutectics after laser zone remelting, which is obviously different with the sintered composite with the same composition (Fig. 1c). Additionally, the interphase spacing in the laser remelted ternary hypoeutectic composite is obviously smaller than that of the Al_2O_3/YAG binary eutectic (Fig. 1f), but is larger than that of the ternary eutectic (Fig. 1d) grown at the same conditions. Similar phenomena are also observed by Waku et al. [28] in directionally solidified alumina-based composites prepared by Bridgman method. This reveals that the addition of the third phase (ZrO_2) can effectively lead to a refinement of microstructure. Consequently, an important improvement in property can be obtained [17,20].