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Phase transformation behavior in air plasma sprayed yttria stabilized zirconia coating

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ABSTRACT

Yttria stabilized zirconia has been widely applied as thermal barrier coatings for high temperature heat insulation. However, prepared by air plasma spraying, YSZ TBCs suffer from severe phase separation of metastable T′ phase into thermodynamic equilibrium T and C phases, which consequently leading to cracking and delamination of the coatings. In the present research, the kinetics of T′ → T + C reaction was experimentally investigated. By AVARAMI equation fitting, the reaction order and apparent activation energy were obtained to be 0.7–1.4 and 515 ± 32 kJ/mol, indicating a complex crystallization and grain growth controlling mechanism of the phase degradation process. Combination effect of temperature and time on the reaction was normalized by Hollomon-Jaffe Parameter. The results provide a reasonable prediction of the phase evolution, as well as the related thermal and mechanical properties of TBCs during long term application.

1. Introduction

Successful thermal protection of high temperature components in modern gas turbines has been accomplished by applying ceramic thermal barrier coatings (TBCs) onto the superalloy components surface to prolong their life time. The coatings are designed to work for long time in severe conditions including high temperature, high pressure, large thermal gradient, and corrosive environment. The primary choice for commercial TBCs are 6–8 wt% yttria stabilized zirconia (YSZ) material with low thermal conductivity (2.3 W/m*K at 1000 °C) [1,2] which provides excellent mechanical properties, thermal insulation and oxidation protection in current applications.

The refractory nature of YSZ TBC materials requires high-energy processing capability, hence air plasma spraying (APS) or electron beam physical vapor deposition (EB-PVD) methods are primarily used in TBC preparation [1,2]. Recently, new processing technologies such as suspension and solution plasma spraying (SPS) and plasma spray physical vapor deposition (PS-PVD) have also been explored [2]. All these methods undergoing a non-equilibrium process result in a metastable tetragonal prime (T′) phase of YSZ. This meta-stable single-phase structure exhibits good comprehensive mechanical properties at both room and high temperatures, mainly due to the ferroelastic toughening mechanism of the T′ phase [3,4]. However, according to the ZrO_2-Y_2O_3 phase diagram in Fig. 1(a) [5], the equilibrium phase composition at the turbine operation temperature range is Y-poor tetragonal (T) + Y-rich cubic (C) phase, and the T′ phase is susceptible to decomposition into T + C during operation.

The newly precipitated T phase, unfortunately, is a transformable phase; a martensitic tetragonal/monoclinic (T/M) phase transformation will occur at lower temperature, accompanied with large volume expansion (5~7 vol%), leading to cracking and spallation of the TBCs. This is the main failure reason in zirconia-based ceramics [6]. Fig. 1(b) [5] shows the crystal cells of the T′, T, C and M phase. Each crystal cell can be regarded as a quasi-fluorite structure with different Y^{3+} ion doping, tetragonality c/a, and oxygen ion displacement. The similarities in structure of them are not enough to guarantee similar properties, so the excellent performance of the T′ phase YSZ TBCs deteriorates due to high temperature phase transformation. This is one of the reasons why the long-term application of YSZ-based TBCs is not practical at higher operation temperature [7].

The aim of developing the next generation gas turbines is to further increase the inlet temperature for higher efficiency. This calls for both a better understanding of the phase transformation and methods to increase the T′ phase stability of YSZ at high temperature. There are several methods to study the phase transformation process in YSZ ceramic such as TEM (Transmission

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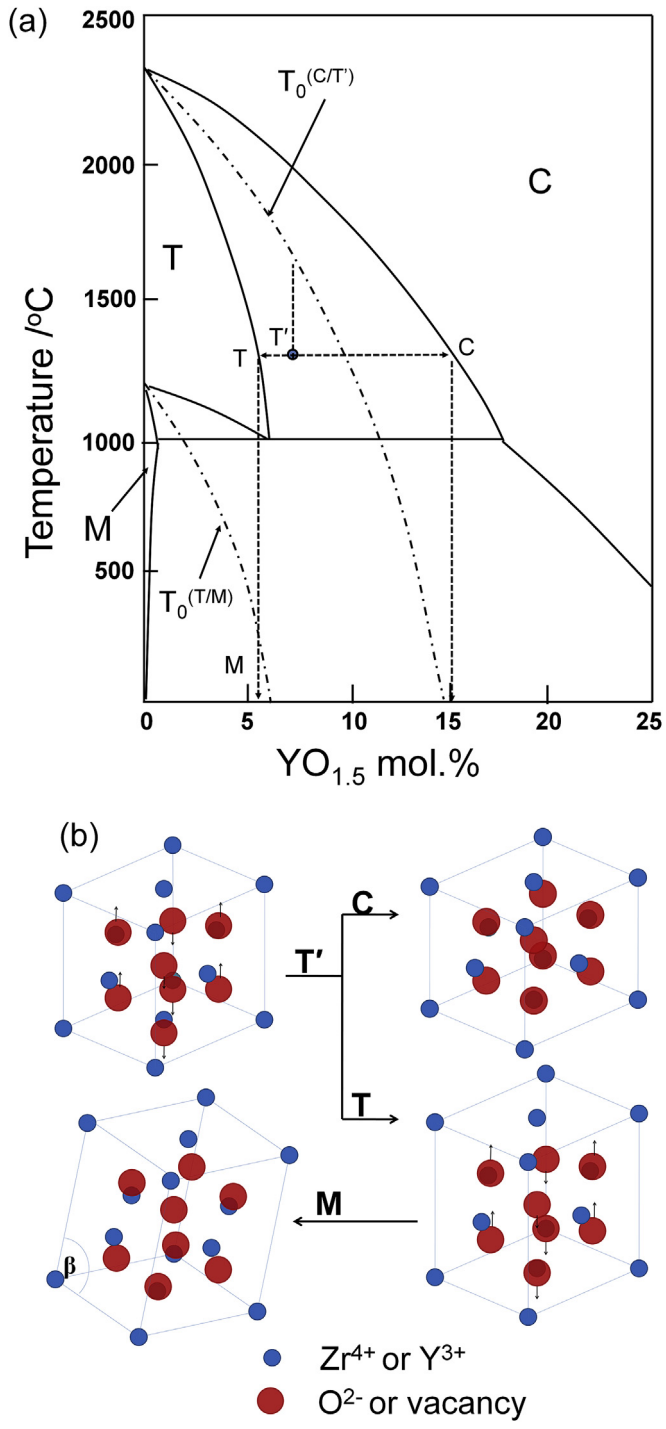

Fig. 1. (a) Phase diagram of ZrO_2-Y_2O_3 system at zirconia-rich corner; (The dash-dot lines indicate the non-equilibrium diffusionless phase transformation from C to T′ and from T to M, respectively.) [10] (b) Crystal cell structure of different phases in YSZ [5]; 1) T′ phase, $P4_2/nmc$, c > b = a, β = 90° oxygen displacement; 1) T′ phase, $P4_2/nmc$, c > b = a, β = 90° oxygen displacement; 2)T phase, $P4_2/nmc$, $c_T > c_{T'} > b = a$, β = 90°, larger oxygen displacement, lower Y^{3+} concentration and oxygen vacancies; 3) C phase, Fm3m a = b = c, β = 90°; 4)M phase, $P2_1/c$, c > b>a, β = 99.194°.

electron microscopy) [8], Raman spectroscopy [9], EPMA (Electron Probe X-ray Micro Analysis) [5] and XRD (X-Ray Diffraction) [10,11]. Each of these has its specific advantage and in complementary to the other methods. TEM can provide lattice parameters, atom arrangement and stabilizer segregation at grain boundaries with high resolution, but it lacks quantitative information of phase composition. Raman spectroscopy is a non-destructive method to provide chemical bond structure, lattice distortion and internal stress information, and is especially sensitive to the monoclinic structure. However, long-range structural information is absent in Raman spectrum. EPMA can be used to establish a microstructure-phase relationship during phase transformation, but no lattice information is provided. As for XRD, the most widely used and effective method in YSZ TBC characterization, it can provide structural information for a longer range than Raman and quantitative phase composition information could also be provided by XRD intensity [11]. By using these techniques, a general understanding of the T′ → T + C transformation can be shaped, but is still far from satisfactory considering the complexity and significance of the information carried by XRD. For example, the mechanism of phase transformation is still not clear and debates between the nucleation & grain-growth model and spinodal decomposition model still exists [5,10]. To increase the phase stability of YSZ, some researchers tried to add a third or multiple stabilizers into YSZ [12–16], but so far, none of them works effectively without sacrificing mechanical properties of YSZ TBCs [3].

Without a complete understanding of the mechanism of the T′ → T + C transformation, it will be nearly impossible to increase the phase stability of YSZ TBCs for higher temperature applications. This is a part of our study on the phase transformation of YSZ TBCs and in this work, we focus on a better understanding of the T′ → T + C transformation from thermodynamics and kinetics aspects by analyzing the phase transformation process at six high temperature thermal treatment conditions with the help of quantitative XRD analysis. And we will try to establish the relationship between phase transformation and the thermal condition of YSZ TBCs during operation.

2. Sample preparation and phase characterization

Eight weight percent (8 wt%) yttria stabilized zirconia ceramic TBC samples with a thickness of 2 mm were prepared by an air plasma spraying unit (GTV-MF-P-HVOF-K-ARC, F6 spraying gun, GTV, Betzdorf, Germany)) on iron-based metal substrates (20 mm × 40 mm × 1.5 mm). The coatings were cut into 10 mm × 10 mm pieces after careful removal from the substrates by 3–5 times of thermal shock cycling. The thermal shock condition is as low as 500 °C for 5min each time which would not affect the meta-stable T′ phase. The removal step was essential since the metallic substrates would be oxidized in air and iron ions would contaminate YSZ coating due to subsequently diffusion at high annealing temperature (>1100 °C). The high temperature aging experiment was conducted in air atmosphere using a muffle furnace (LHT04/17, Nabertherm, Germany) at six different temperatures, 1100 °C, 1150 °C, 1200 °C, 1250 °C, 1300 °C and 1350 °C. This temperature range covers the working temperature of the current state-of-art TBCs and that of TBCs in next generation gas turbines. Length of aging time was selected according to the annealing temperature, ranging from several hours (1350 °C) up to seven hundred hours (1100 °C). Annealed samples were then fetched out of the furnace at designed times, which was followed with air cooling. Sample surface were machined and polished by diamond polishing slurry (minimum particle size: 3 μm) to reduce experimental errors in the following step.

High-resolution XRD patterns of all samples were recorded from 72.5° to 75.5° at a scanning speed of 0.3°/min using an X-ray Diffractometer (D/max-2500 High-power Multi-crystalline X-ray Diffractometer, Rigaku, Japan) equipped with Cu $K_{\alpha 1}$ monochromator to exclude interfering peaks from Cu-K_{a2} X-ray.

Quantitative XRD analysis of multi-phase crystalized material samples can be applied to obtain the phase composition. According