



Contents lists available at ScienceDirect

Journal of the European Ceramic Society

journal homepage: www.elsevier.com/locate/jeurceramsoc

Original Article

On the Yttrium Tantalate – Zirconia phase diagram

Mary Gurak¹, Quentin Flamant, Laetitia Laversenne², David R. Clarke**John Paulson Harvard School of Engineering and Applied Sciences, Harvard University, Cambridge, MA 02138, USA*

ARTICLE INFO

Keywords:

Phase equilibria
Zirconia
Yttrium tantalate
Phase transformations
Thermal barrier coatings

ABSTRACT

The phase diagram for the YTaO₄-ZrO₂ quasi-binary has been determined up to 1600 °C. There are three distinct compositional regimes: an extensive YTaO₄ solid solution, an extensive ZrO₂ solid solution and a two-phase intermediate region. The addition of ZrO₂ to YTaO₄ decreases the *M*-*T* transition temperature almost linearly from 1426 °C to approximately 450 °C at the solubility limit (~28 mol% ZrO₂), and then remains constant until the ZrO₂ solid solution phase boundary is reached. Within the intermediate region, there exists an extensive two-phase tetragonal (*T* + *t*) phase field above the *M*-*T* transformation temperature. The transformation exhibits no hysteresis on heating and cooling but nonetheless there is a distribution with temperature in the mass fraction of the monoclinic and tetragonal phases so no unique transformation temperature can be identified. No other high temperature phases were observed but it is suggested that a higher temperature solid solution phase is likely above 1700 °C, based on the similarity in crystallographic relationship between the two tetragonal solid solution structures.

1. Introduction

It is widely recognized in the turbine materials community that the engine efficiency, whether for power generation or propulsion, increases with the high-temperature turbine inlet temperature [1]. Over the years since the first gas turbines were built there have been several developments that have enabled designers to meet the challenge of increasing temperatures. These include the development of single crystals of metal alloy compositions capable of higher temperature creep and fatigue resistance [2], the use of ever more complicated internal cooling of the turbine blades [3,4] and the introduction of thermal barrier coatings of the blades and vanes [1,5]. Since the introduction of thermal barrier coatings, the material of choice has been, and continues to be, yttria-stabilized zirconia in its meta-stable tetragonal-prime (*t'*) phase [6,7]. This has a combination of attractive properties: low thermal conductivity [8], high fracture toughness [9] and ease of deposition over complex shapes [7]. Increasingly, as the turbine inlet temperature is being raised further, some of the very highest temperature limitations of this oxide are being recognized. Principal amongst these are the observations that the meta-stable *t'* phase undergoes a slow but temperature-dependent, kinetically-limited transformation to a thermodynamically stable mixture of cubic and tetragonal phases, the latter of which can transform to monoclinic zirconia on cooling [6,10]. The concern with this transformation is that

the large accompanying volumetric expansion can cause cracking and accelerated failure of the coating. The kinetics of the meta-stable tetragonal conversion are well described by the Larson-Miller relation even though the actual exponent has been found to depend on the actual measurement made to measure the transformation [6,11]. For engine designers, the importance of the Larson-Miller fitting is that it allows them to estimate the combination of temperatures and times at temperature before the transformation will occur.

Although it is now known that there are many oxides [12] that are capable of withstanding higher temperatures than the meta-stable form of yttria-stabilized zirconia and also have lower thermal conductivity, they lack any intrinsic toughening mechanism. Many can also be difficult to deposit, especially at high rates on curved surfaces. More limited in number are the high-temperature oxides that undergo some form of displacive or martensitic phase transformation at high temperatures. One of these is yttrium tantalate (YTaO₄) which early reports suggested had a tetragonal-to-monoclinic transformation at about 1450 °C [13,14], considerably higher than the corresponding transformation temperature in pure zirconia (~1060 °C). Based on several literature reports that the tetragonal form of zirconia can be stabilized by equal concentrations of Y³⁺ and Ta⁵⁺, this work reports investigations of the pseudo-binary phase diagram between YTaO₄ and ZrO₂, extending previous studies [15] of the phase transformations at the YTaO₄ end of the diagram. A further attractive feature of this system is that

* Corresponding author.

E-mail address: clarke@seas.harvard.edu (D.R. Clarke).¹ Now at: Pratt and Whitney Aircraft, Hartford, CT, USA.² Now at: Institute NEEL CNRS, 38042 Grenoble, France.<https://doi.org/10.1016/j.jeurceramsoc.2018.03.012>Received 21 January 2018; Received in revised form 9 March 2018; Accepted 9 March 2018
0955-2219/ © 2018 Elsevier Ltd. All rights reserved.

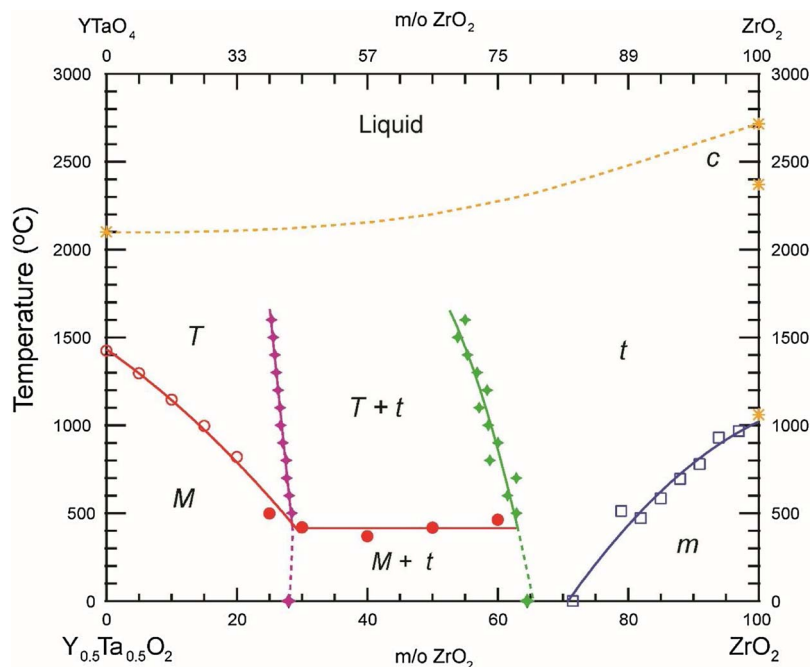


Fig. 1. Phase diagram along the YTaO_4 - ZrO_2 quasi-binary. The filled markers correspond to this work, and the open markers are data from literature including our own [1,5,8]. The room temperature solubility limits are marked by the vertical dashed lines. For convenience, the compositions are indicated both in terms of molar mixtures of YTaO_4 and ZrO_2 (top) and in terms of constant number of cations and anions (bottom), the notation used in this work.

fully-dense compositions along this join have considerably lower thermal conductivities than that of 8YSZ [16].

The majority of previously reported phase diagram studies have focused on the zirconia-rich end of the YTaO_4 - ZrO_2 diagram, Fig. 1. Complementary studies by XRD [17–19], Raman [20] and dilatometry in this compositional region indicate that co-doping with equal amounts of Y^{3+} and Ta^{5+} decreases the tetragonal-monoclinic (t - m) transformation temperature monotonically with concentration up to about 25 m/o YTaO_4 substitution into ZrO_2 . Over this compositional range the tetragonal phase can be retained on cooling depending on the grain sizes and cooling rates otherwise it transforms martensitically to a monoclinic zirconia solid solution [20]. At the other end of the pseudo-binary, YTaO_4 , previous studies [15] of the phase stability show that ZrO_2 can substitute into YTaO_4 up to at least a concentration of 25 m/o ZrO_2 . Over this compositional range the T - M phase transformation temperature decreases from 1426 °C for pure YTaO_4 down to ~500 °C [15]. Most significantly for potential toughening, the transformation is purely displacive over the entire solid solution [15,21]. Intriguingly, the XRD studies of the tetragonal-monoclinic (T - M) transformation in the single-phase YTaO_4 solid solution region also indicate that a proportion of the tetragonal (T) phase can be retained on cooling of the powders to room temperature [15]. This is analogous to the retention of the t' tetragonal prime phase on cooling at the ZrO_2 end of the diagram.

Preliminary microstructural observations and room temperature XRD measurements suggest that even though both the YTaO_4 solid solution and the ZrO_2 solid solutions are tetragonal, there exists a two-phase co-existence region between them. The work presented in this contribution describes the determination of the compositional limits of the two solid solutions and the phase transformation behavior in the two-phase region. As the kinetics in this system are very slow, the emphasis in this work has been on X-ray measurements primarily carried out on powders, rather than bulk samples, in air at high temperatures. The slow grain growth kinetics in this material system is evident from the grain size in dense samples even after being held at 1600 °C for 40 h, as shown in Fig. 2.

To avoid confusion, the tetragonal and monoclinic forms of the zirconia-solid solution phases are indicated by the lower case letters t - and m -, respectively, whereas those of the yttrium-tantalate solid solution are indicated by the upper case letters T - and M -. Compositions are expressed in this work in terms of mole percent of single cation

formula units, namely as $\text{Y}_{(1-x)/2}\text{Ta}_{(1-x)/2}\text{Zr}_x\text{O}_2$ so the terminal phases are $\text{Y}_{0.5}\text{Ta}_{0.5}\text{O}_2$ ($x = 0$) and ZrO_2 ($x = 1$).

2. Experimental details

The majority of our studies have been performed on fine powders prepared by the reverse co-precipitation method [15,22,23]. Mixed cation solutions were prepared from zirconium oxy-nitrate hydrate (> 99%) and yttrium nitrate hexahydrate (> 99.8%) aqueous solutions mixed with tantalum chloride (99.99%) solutions in ethanol, with the concentrations calibrated using the gravimetric method. The mixed solution was then added drop-by-drop into an ammonium hydroxide solution at an initial pH of 11.2 at room temperature, stirring the whole time and with the pH maintained above 10.6. White precipitates were formed and then separated by centrifugation. These were subsequently washed three times, twice with deionized (DI) water and once with ethanol, before being dried overnight. Finally, the powders were calcined in air at 700 °C for 2 h to create molecularly mixed metal oxides. Based on DSC studies, the powders crystallize to the monoclinic-prime phase at temperatures dependent on the ZrO_2 content. These then transformed to the equilibrium phases at higher temperatures, as confirmed by Raman spectroscopy. For the X-ray diffraction (XRD) studies presented in this work, all the calcined powders were first heated at 1600 °C for 40 h so that they became tetragonal. These conditions were selected based on preliminary measurements.

Phase identification was performed by X-ray diffraction using different facilities. The highest angular resolution measurements at room temperature were made at the Advanced Photon Source (APS) at Argonne National Laboratory (in Argonne, Illinois, USA) using the 11-BM mail-in program. Prior to making these measurements, the calcined powders underwent different heat treatments before being ground with a mortar and pestle and then passed through a 325 mesh sieve. The X-ray wavelength was 0.459981 Å (27 keV).

High temperature diffraction studies of the 25 mol% ZrO_2 and 30 mol% ZrO_2 compositions were performed up to 1000 °C using a PANalytical X'Pert PRO diffractometer equipped with an Anton Paar HTK1200N furnace at the MIT Center for Materials Science and Engineering. The patterns obtained on this instrument were analyzed using the Rietveld software as part of the X'pert HighScore Plus analysis package.

Download English Version:

<https://daneshyari.com/en/article/7898252>

Download Persian Version:

<https://daneshyari.com/article/7898252>

[Daneshyari.com](https://daneshyari.com)