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Original Article

# The effect of zirconia substitution on the high-temperature transformation of the monoclinic-prime phase in yttrium tantalate

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## ABSTRACT

Amorphous yttrium tantalate, as well as solid solutions containing zirconia, transform on heating to a monoclinic-prime phase and then, with further heating, to a crystalline tetragonal (T) solid solution phase at  $\sim 1450^\circ\text{C}$ . On subsequent cooling the tetragonal phase converts by a second-order displacive transformation to a different monoclinic phase not to the monoclinic-prime phase. On subsequent reheating and cooling, the phase transformation occurs between the monoclinic (M) and tetragonal phases, and the monoclinic-prime phase cannot be recovered. The limit of zirconia solubility in both the monoclinic-prime and monoclinic phases lies between 25 and 28 m/o  $\text{ZrO}_2$ , consistent with previous first-principles calculations. The monoclinic-prime phase is stable up to at least  $1400^\circ\text{C}$  for 100 h for zirconia concentrations from 0 to  $\sim 60$  m/o  $\text{ZrO}_2$ . This temperature exceeds the temperature of the equilibrium *M-T* phase transformation suggesting that the monoclinic-prime phase transforms directly to the tetragonal phase by a reconstructive transformation and is unaffected by the zirconia in solid solution.

## 1. Introduction

Compositions lying between  $\text{YTaO}_4$  and  $\text{ZrO}_2$  have gained considerable interest [1–4] recently because their combination of low thermal conductivity [1,2] and high temperature stability makes them attractive for high-temperature applications, such as thermal barrier coatings [3]. Furthermore, at the  $\text{YTaO}_4$  end of the pseudo-binary with  $\text{ZrO}_2$ , the high-temperature transformation between the equilibrium tetragonal (T) and monoclinic (M) phase is unusual because it is a second-order, displacive transformation [5]. The transformation in pure  $\text{YTaO}_4$  occurs at  $1426^\circ\text{C}$  and, intriguingly, additions of  $\text{ZrO}_2$  up to  $\sim 28$  mol percent (m/o)  $\text{ZrO}_2$  do not disrupt the transformation although they do decrease the transformation temperature [5]. Based on ionic size and preferred coordination, the substitution of zirconia into  $\text{YTaO}_4$  is unexpected. Furthermore, our studies of the monoclinic-tetragonal phase transformation were of materials that had been homogenized and heated into the tetragonal phase. This is significant, since in the older literature, investigating the niobates and tantalates as phosphor hosts, another monoclinic phase, termed monoclinic-prime (M-prime), has been documented [6–9] to form preferentially at low temperatures from both amorphous precursors and mixed oxides and is resistant to transformation to the monoclinic phase (M) until high temperatures. It has also been suggested in at least one paper that the

M-prime phase of  $\text{YTaO}_4$  is a thermodynamically stable phase up to  $\sim 1450^\circ\text{C}$  [9]. However, recent first-principles computations suggest that the M-prime phase is not an equilibrium phase of either pure  $\text{YTaO}_4$  or with  $\text{ZrO}_2$  substitution [10]. None of these papers, or others in the literature, report on the substitution of tetra-valent ions in  $\text{YTaO}_4$ .

In this work, we use a combination of techniques, including *in-situ* Raman measurements as a function of temperature, to characterize the phase transformation behavior of amorphous  $\text{YTaO}_4$  and solid solutions with zirconia to explore the effect of zirconia substitution. As the transformation behavior between the monoclinic and tetragonal forms of  $\text{YTaO}_4$ , as well as the effect of zirconia substitution on this high temperature transformation, has been studied in detail [5], the emphasis of the current contribution is on the intermediate, irreversible transformations when the starting material is an amorphous material containing zirconia in solid solution. We will also address the question of under what conditions the monoclinic phase forms since, as far as we are aware, based on the literature [11] and our own work, the monoclinic phase cannot be formed directly from the M-prime phase and can only be formed on cooling from the tetragonal phase, even when molecularly mixed precursors are used.

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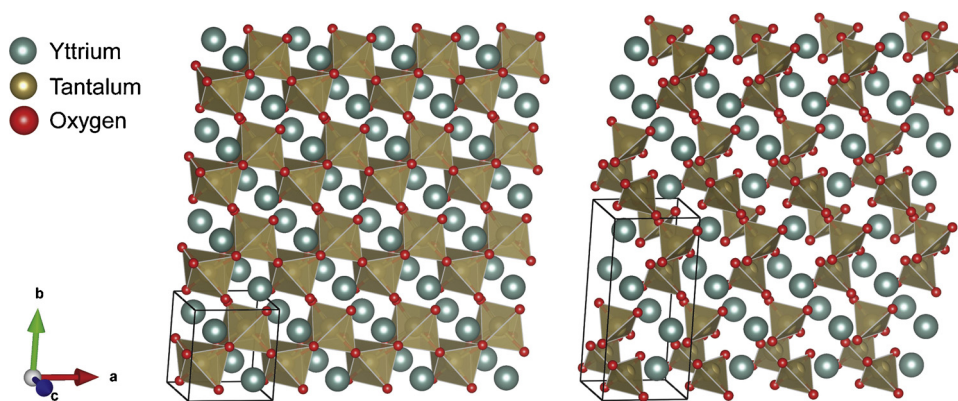


Fig. 1. The crystal structures of the two monoclinic forms of  $\text{YTaO}_4$ , *M*-prime (left) and monoclinic (right). A unit cell is delineated in black of each phase. The alignment of the distorted  $\text{TaO}_6$  units to form chains and the alternating planes of Y and Ta in the *b*-*a* plane of the *M*-prime structure can be seen.

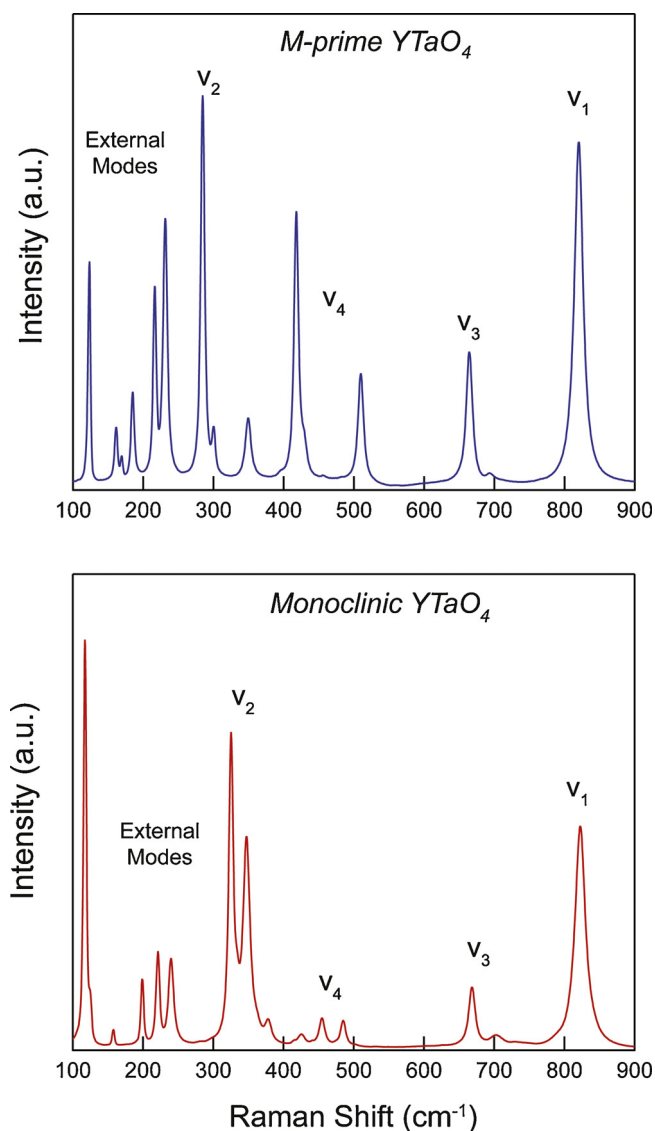


Fig. 2. Raman spectra of the monoclinic-prime and monoclinic forms of  $\text{YTaO}_4$ .

## 2. Experimental details

### 2.1. Material synthesis and preparation

Powders of  $\text{YTaO}_4$  and mixtures with  $\text{ZrO}_2$  with overall composition

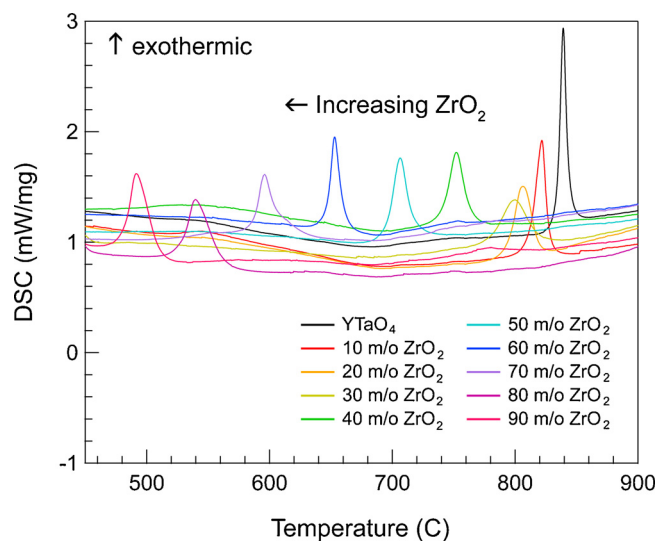


Fig. 3. DSC measurements showing the crystallization temperatures of compositions along the  $\text{YTaO}_4$ - $\text{ZrO}_2$  compositional line. The measurements were performed on powder samples produced by co-precipitation after drying.

given by the atomic relationship  $\text{Y}_{(1-x)}\text{Ta}_{(1-x)}\text{Zr}_{2x}\text{O}_4$  were synthesized using the reverse co-precipitation method [12] to obtain molecularly mixed powder as has been described previously [2,13]. Precursor solutions of zirconium oxynitrate hydrate (> 99%) and yttrium nitrate hexahydrate (> 99.8%) were prepared by dissolving the powders in deionized (DI) water, while tantalum chloride (99.99%) was dissolved in ethanol. The precursor solutions were calibrated using the gravimetric method, with a target concentration of 0.4 mmol/g, and subsequently mixed in the proper proportions to yield the desired composition. The mixed solution was then inserted drop by drop into a 2.5 M ammonium hydroxide solution,  $\text{NH}_4\text{OH}$ , with an initial pH of 11.2 at room temperature, and the pH was maintained above 10 during the process. This pH was chosen based on the insolubility ranges of the individual elements as represented by their Pourbaix diagrams.

The resulting white precipitates were then separated by centrifugation and subsequently washed three times, twice with DI water and once with ethanol, before being dried overnight at 70 °C. Finally, the powders were calcined in air for 2 h to create molecularly mixed metal oxides.

### 2.2. Characterization

Both Raman and X-ray diffraction were used to characterize the materials. Raman spectroscopy was chosen as both the *M* and *M*-prime

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