Stress Measurement of Thin Film Oxide on The Zirconium Metal Using Steam Beam

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1. Introduction

It is well-known that tetragonal ZrO_2 forms in the metal-oxide interface in the early stage of zirconium oxidation that is protective against further oxidation. However, as the oxide grows the stress built up during the oxidation process relieves then the tetragonal phase turns into the monoclinic phase which is non-protective and stable at low stress. Therefore, it is believed that the zirconium oxidation kinetics depends on the phase transformation that take place at 3.0 GPa according to the earlier works. In other words, if the transformation is related by holding the oxide stress above the threshold stress, the kinetics would be slow. Nevertheless, so far no one has succeeded to measure the stress of thin film oxide above 3.0 GPa. In this study, the stress has been measured using steam beam apparatus successfully above the threshold stress level.

2. Methods and Results

2.1 Experimental

In-situ bend test apparatus using steam beam is designed Figure 1. Here in the apparatus, Oxidation reaction occurs only the surface exposed to the steam beam. In order to avoid the oxidation of the other side of specimen, whole chamber is evacuated down to ultra-high vacuum (down to 10^{-6} Torr).

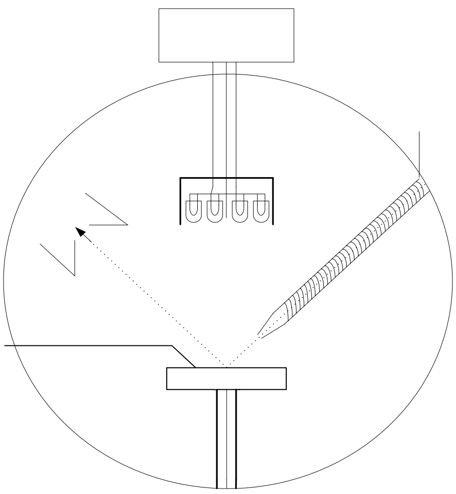

Figure1. In-situ bend test apparatus using steam beam in ultra-high vacuum.

The oxide thickness is measured with weight gain measurement and the curvature of the single side oxidized specimen is measured with spherometer. Specimen is thin film zirconium foil whose thickness is 40 ㎛ and diameter is 20mm. Only single surface of specimen exposed to the steam beam oxidizes at 400℃ which is attained by halogen lamp.

2.2 Stress measurement

Basically the measurement technique used in this study is based on the curvature build-up during the single side oxidation process. The stress build-up can be directly evaluated according to the Stoney’s formula.

$$\sigma_f = \frac{E}{1-\nu}\frac{t_s^2}{6t_f R} \quad (1)$$

Where ,
E : Young' Modulus of metal   v : Possion ratio
R : radius of curvature t_f : oxide thickness
t_s : metal thickness

Measured stress from the curvature estimation are plotted as a function of thin oxide film thickness in Figure 2.

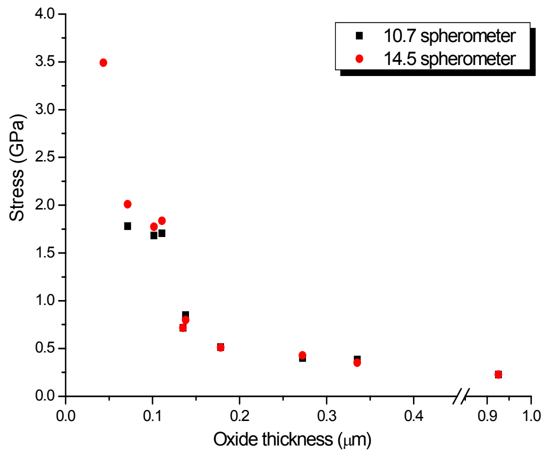

Figure 2. Measured stress from the curvature estimation with thin oxide film thickness.

As shown in the figure, the stress in the oxide increases as the thickness decreases and the highest stress measured in this study is 3.4 GPa which is higher than any other previous work. In fact, the measured correlation between the stress and the oxide thickness is in good agreement with the theoretical prediction. Taking into consideration that the measured stress is the average stress of whole oxide, not in the interface. Therefore, these results reveal that stress built up in the interface can exceed the threshold value, which supports the phase transformation theory.

3. Conclusion

In this study, stress of thin film oxide grown on the zirconium metal has been measured using steam beam in the ultra-high vacuum system. The highest stress measured in the test is 3.4 GPa with about 0.04 ㎛ oxide thickness, which can support the phase transformation theory in the zirconium oxidation kinetics. In the following study, the direct measurement of phase transformation at the threshold stress using Raman spectroscopy and the effect of hydride precipitation on the transformation will be carried out.

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