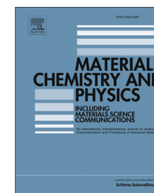
Materials Chemistry and Physics xxx (2015) 1–16

Contents lists available at ScienceDirect

Materials Chemistry and Physics

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



Thermal stability and thermal expansion behaviour of ZrO_2/Y_2O_3 multilayers deposited by pulsed laser deposition technique

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HIGHLIGHTS

- ZrO_2/Y_2O_3 multilayers were deposited by pulsed laser deposition technique.
- Formation of tetragonal phase of ZrO_2 and cubic phase of Y_2O_3 were observed.
- The multilayers films show good thermal stability upto temperature 1373 K.
- The coefficient of thermal expansion (CTE) of t-ZrO_2 decreases with increase in ZrO_2 layer thickness.

ARTICLE INFO

Article history:
Received 9 April 2014
Received in revised form
10 April 2015
Accepted 15 June 2015
Available online xxx

Keywords:
Multilayer
Physical vapour deposition
X-ray diffraction
Thermal expansion

ABSTRACT

Multilayers of ZrO_2/Y_2O_3 were prepared by pulsed laser deposition technique with variation in the ZrO_2 layer thickness from 5 to 30 nm keeping the Y_2O_3 layer thickness constant (~10 nm). The stability, phase evolution and thermal expansion behaviour of the multilayers were analyzed by high temperature x-ray diffraction technique, in the temperature range of 300–1373 K. Unlike the single layer of ZrO_2 film, which shows a mixture of tetragonal and monoclinic phase, the ZrO_2 layers in multilayers show tetragonal phase in case of all the multilayers investigated in the present work. The values of coefficient of thermal expansion (CTE) decrease with increase in the ZrO_2 layer thickness. The CTE of both ZrO_2 and Y_2O_3 are found to be influenced by their mutual solubility as well as due to interdiffusion of these oxides taking place along the interfaces of the multilayers, especially during high temperature heat-treatment.

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1. Introduction

At ambient pressure condition, zirconium oxide (ZrO_2) exists in three different phases such as monoclinic (m), tetragonal (t) and cubic (c). Each phase of ZrO_2 has its own applications in different fields, for examples m-ZrO_2 is used in biological applications [1], t-ZrO_2 in thermal barrier coating (TBC) [2] and c-ZrO_2 in sensors [3] and solid oxide fuel cells (SOFC) [4]. A common method of stabilization of the high temperature phases of ZrO_2 is by doping with suitable transition metal oxides (Y_2O_3 and Al_2O_3) [5,6] or by decreasing the crystallite sizes below a critical value [7–9]. Recently, formation of multilayers is another interesting method, which is being followed by researchers to stabilize the high temperature phases of ZrO_2 at room temperature [10–12].

Coefficient of thermal expansion (CTE) of films is considered to be one of the important parameters for the selection a material for a particular application. Materials used in solid oxide fuel cells (SOFC), thermal barrier coatings (TBC), microelectronics and micromechanical systems demand low thermal expansion mismatch in order to reduce thermal degradation and cracking during the operation. The common techniques that are being used to measure the CTE values for bulk materials are mechanical dilatometry [13], optical interferometry [14] and diffraction techniques [15]. In case of the thin film structures, these methods cannot be used because of lack of accuracy in the measurements due to smaller thickness. Measurement of the CTE values of a thin film can be carried out by HTXRD [16], ellipsometer [17] and micro-machined cantilever techniques [18]. Out of all the techniques high temperature X-Ray diffraction (HTXRD) finds its importance in the determination of CTE values due to the following advantages: (i) smaller size of sample can be studied and (ii) the expansion along

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http://dx.doi.org/10.1016/j.matchemphys.2015.06.033
0254-0584/© 2015 Published by Elsevier B.V.

Please cite this article in press as: M. Mishra, et al., Thermal stability and thermal expansion behaviour of ZrO_2/Y_2O_3 multilayers deposited by pulsed laser deposition technique, Materials Chemistry and Physics (2015), http://dx.doi.org/10.1016/j.matchemphys.2015.06.033

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different crystallographic directions can also be determined. However, HTXRD is suitable only for crystalline films and is an important technique to study the thermal stability, phase evolution as well as inter-diffusion phenomenon occurring during the heat-treatment.

Swamy et al. [19] have studied the structural transitions and thermal expansion behaviour of Y_2O_3 from room temperature to its melting point. The axial thermal expansion of ZrO_2 has been studied by Patil et al. [20] and they have reported the CTE values to be 11.6 and 16.08 × 10^{-6} K^{-1} along *a* and *c* axes, respectively for t-ZrO_2.

Although phase stability and calculation of the CTE values for single oxide layer have been reported by many authors, a very few research papers have been published on multilayer systems. Barshilia et al. [21] have deposited TiN/CrN multilayers by sputtering technique and studied the thermal stability of the multilayers by HTXRD. Balakrishnan et al. [10,11] have analyzed the thermal stability and thermal expansion behaviour of ZrO_2/Al_2O_3 and CeO_2/ZrO_2 multilayer films deposited by pulsed laser (PLD) technique. Barshilia et al. [12] have prepared ZrO_2/Al_2O_3 and ZrO_2/Y_2O_3 by sputtering technique and analyzed the microstructural and nano-mechanical properties of the films. However, deposition of ZrO_2/Y_2O_3 multilayer films by PLD technique and study of their thermal stability and CTE of the multilayers of ZrO_2/Y_2O_3 as a function ZrO_2 layer thickness have not been reported so far. The present study reports the preparation of multilayer films of ZrO_2/Y_2O_3 on Si (100) substrates with different ZrO_2 layer thickness and the effect of ZrO_2 layer thickness on stability of multilayer structure as well as the coefficient of thermal expansion (CTE). The films were characterized by high temperature X-ray diffraction (HTXRD) and high resolution transmission electron microscopy (HRTEM) techniques.

2. Experimental procedure

Multilayers of ZrO_2/Y_2O_3 were prepared from sintered ZrO_2 and Y_2O_3 targets by pulsed laser deposition technique. The details of the target sintering methods, their characterizations and substrate cleaning methods were already reported in previous publications [22,23]. A KrF excimer laser (Lambda Physik Compex-205, 248 nm wavelength) was used as the energy source to evaporate the targets for multilayer deposition. The multilayer films were prepared at substrate temperature of 300 K and oxygen partial pressure of 2 Pa. The details of the deposition parameters were listed in Table 1. The layer thickness of ZrO_2 was varied from 5 to 30 nm, whereas the Y_2O_3 layer thickness was kept at 10 nm. The total thickness of the multilayer films was kept constant by varying the bilayer thickness (modulation wavelength) (Table 1). The total number of bilayers for the ZrO_2 layer thickness of 5, 10, 20, and 30 nm were 65, 50, 35 and 25, respectively. Single layers of ZrO_2 and Y_2O_3 were also deposited on Si (100) substrates at the substrate temperature of 300 K and oxygen partial pressure of 2 Pa to optimise the deposition conditions necessary for the growth of the multilayers.

HTXRD experiments of the single as well as multilayer samples were carried out using an X-ray diffractometer (INEL XRG-3000, France) attached with a high temperature arrangement (Buhler HDK 2.4) at an incident angle of (ω) of 5°. Films deposited on Si (100) substrates were kept on a tantalum sample holder and in-situ heated in the temperature range of 300–1373 K. To maintain a uniform temperature, the substrate holder was radiatively heated by surrounding heater made up of tantalum. The temperature was measured using a W–Re thermocouple with ±2 K tolerance. During the heat-treatment schedule, a heating rate of 10 K/min was followed and data were recorded for a time period of 15 min after 3 min of stabilization time using a curved position sensitive detector in the 2Θ range of 10°–100°. After the measurements, the samples were cooled at a rate of 50 K/min to room temperature. The measurement of CTE values by HTXRD is only applicable for crystalline films [24], and also it is a well known that all the thin film structures are always associated with internal stress and defects [25,26], Since the latter factors also known to contribute to the change in the lattice parameters as a function of temperature, Lebrun et al. [27] have suggested that annealing of the film at suitable temperatures is necessary to determine the coefficient of thermal expansion more accurately. In the present study, all the multilayer films were heat-treated in two steps. The first heat-treatment was carried out to relieve stress and to form a crystalline film and the second heat-treatment was carried out to collect the data on lattice parameter in order to determine the thermal expansion values. The heat-treatment steps and range of temperature is the same in both the heat-treatment steps.

From the HTXRD pattern, the changes in the lattice parameter and phase stability were monitored and the lattice parameters were calculated using “Unit ell” program with second order polynomial fitting [28]. From the lattice parameter data, average linear coefficient of thermal expansion (α_{av}) as well as instantaneous coefficient of thermal expansion (α_{inst}) values were calculated using equations 1 and 2, respectively [29,30].

$$\alpha_a = \frac{1}{a_{RT}}\left[\frac{a_T - a_{Rt}}{T - RT}\right] K^{-1} \tag{1}$$

$$\alpha_{inst} = \frac{1}{a_T}\left[\frac{\partial a}{\partial t}\right] K^{-1} \tag{2}$$

where α_a is the average thermal expansion coefficient along a axis, a_T and a_{RT} are the lattice parameter values at temperature T and room temperature (RT), respectively.

The heat-treated multilayers were analyzed by high resolution transmission electron microscopy (HRTEM) to analyze the effect of heat-treatment on interface structure. The HRTEM analysis was carried out using a JEOL 2000 EX II (T) transmission electron microscope having a point resolution of 0.19 nm at an operating voltage of 200 kV. For HRTEM cross sectional specimen preparation, the multilayer samples were cut into small cuboids and two such pieces were glued with G1 epoxy with the film-surfaces facing each other. The whole assembly was then fitted into titanium slot grids. This was thinned down mechanically to ~30 μm and subsequently ion-milled to electron transparency in Technoorg Linda IV 4 ion miller. Extreme care was taken to remove the surface damage of the samples induced by the high-energy ion milling by low energy ion milling at 6 kV and 2 mA in a Technoorg Linda IV 4 Gentle Mill. Phase identification was made from high resolution digital images by calculating power spectra from ~10 nm wide crystalline regions and analysing them for inter-planar spacings and angles. From HRTEM images of multilayers, the individual layer thickness, interface structure, inter-diffusion between layers and defects were examined to study the stability of the multilayers across the

Table 1
Experimental details of the multilayer fabrication by pulsed laser deposition.

Parameters	ZrO_2/Y_2O_3
Laser energy	300 mJ/pulse
Repetition rate	10 Hz
Substrate temperature	300 K
Oxygen partial pressure	2 Pa
Deposition rate	Y_2O_3 (8 nm/min) ZrO_2 (5 nm/min)
ZrO_2 layer thickness (nm)	5,10,20,30
Number of bilayers	65, 50, 35 and 25

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