**A Search for Temperature Induced Time-dependent Structural Transitions in
10mol%Sc₂O₃–1mol%CeO₂–ZrO₂ and 8mol%Y₂O₃–ZrO₂ Electrolyte Ceramics**

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Abstract

Both Sc₂O₃-CeO₂-stabilized-ZrO₂ (SCSZ) and Y₂O₃-stabilized-ZrO₂ (YSZ) show similar Young's modulus damping at 175-400 °C and 200-400 °C, respectively, by impulse excitation acoustic technique. The phase transition in SCSZ is considered responsible for the damping; however for YSZ, such a phase transition has never been reported. To clarify the relation of damping and structural transition in these two materials, the time-dependent high temperature stabilities are studied by *in situ* X-ray diffraction, neutron diffraction and Raman scattering during *long-term* annealing of SCSZ at 350 °C and YSZ at 275 °C. The cubic-to-rhombohedral transition in SCSZ is detected. However, the existence of t' phase in

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4 YSZ raw powders is confirmed without significant changes of the cubic structure during isothermal
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6 annealing. It is concluded that the phase transition contributes to the Young's modulus damping in SCSZ,
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9 while the structural transition is excluded from the reason for damping in YSZ.
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14 **Keywords:** Young's Modulus, X-ray diffraction, Neutron diffraction, Scandia and ceria stabilized
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16 zirconia, Yttria stabilized zirconia
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1 Introduction

Zirconia (ZrO_2) based fast ion conductors are well known and broadly used electrolyte ceramics for solid oxide fuel cells (SOFCs), oxygen separation membranes or as oxygen sensors [1,2]. Yttria stabilized zirconia (YSZ) is one of the most explored compositions, where 8 mol% of Y_2O_3 doping provides sufficient oxygen vacancies in fluorite ZrO_2 lattice to ensure high ionic conductivity up to $\sigma = 0.1 \text{ S/cm}$ and stable cubic structure at 1000°C [1,3]. Because of such high oxygen conductivity and excellent phase stability both in oxygen rich and reducing environments, YSZ electrolyte is applicable to high temperature SOFCs. However, if the operation temperature of SOFC is to be decreased to 800°C , the ionic conductivity of YSZ reduces to $\sigma = 0.044 \text{ S/cm}$ [3-5] which becomes insufficient for high SOFC performance due to very high ohmic losses.

Scandia and ceria doped zirconia (SCSZ) provide an alternative electrolyte material, as the oxygen conductivity of SCSZ is much higher than that of YSZ and is equal to 0.116 S/cm at 800°C [5] which is sufficiently high to ensure SOFC efficient performance in this temperature range. Therefore, a number of publications appeared recently, where SCSZ ceramics were studied as a potential electrolyte material at a great extension [6-11].

While high ionic conductivity is of prime importance to ensure SOFC efficient operation, phase and structural stabilities are playing detrimental role for SOFC durability and longevity. The phase stability of YSZ was studied intensively in past [12-14], and it was found the 8mol% Y_2O_3 - ZrO_2 remained cubic from room temperature to 1000°C or above [15]. Study of the phase stability of 10mol% Sc_2O_3 -1mol% CeO_2 - ZrO_2 revealed that ceramic transformed from cubic to β -rhombohedral phase upon heating at $\sim 300^\circ\text{C}$ and then back to cubic structure at 500°C , and that the structure remained

cubic in 500–1000 °C temperature range [8]. Besides phase stability, the mechanical properties of YSZ and SCSZ are of importance to the reliability of SOFCs, as these ceramics exhibit relatively low strength and fracture toughness [16-18].

Upon investigation of mechanical behavior of YSZ and SCSZ at high temperature, it was found that a high damping behavior occurs during the high temperature measurements of Young's moduli of material by impulse excitation techniques in 200–400 °C for YSZ ceramics and 350–500 °C for SCSZ ceramics, respectively [16,18]. In this temperature range it was not possible to measure Young's modulus or by using the dynamic technique, such as the impulse excitation method. As SCSZ ceramics exhibit cubic to rhombohedral phase transition in the same temperature range, it was hypothesized that damping occurs due to atomic rearrangement of lattice and enhanced atomic vibrations during phase transition in 350–500 °C temperature range. At the same time, there was no reported phase transition for YSZ ceramics in 200–400 °C temperature range, but instead, the ion-vacancy enhanced interactions were used to explain the damping at low temperature in YSZ [18-20]. It was hypothesized [21] that an unreported phase transition that might occur during long time annealing of YSZ could be the reason for the damping.

Therefore, the goal of this paper is to investigate the phase stability of both YSZ and SCSZ ceramics during the annealing of the samples at constant temperature to detect if the phase transition from cubic to lower symmetry phase occurs during long-term annealing.

2 Experimental

Commercial 10mol%Sc₂O₃-1mol%CeO₂-ZrO₂ (SCSZ) powders (ScCeSZ-TC, Nextech, USA) and 8mol%Y₂O₃-ZrO₂ (YSZ) powders (TZ-8Y, Tosoh, Japan) were cold pressed into 7 mm × 10 mm × 70 mm bars under 25 MPa. The bars, covered with the same raw powders, were placed in the alumina tray

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3 and sintered at 1550 °C for 5 h with further machining into specimens with the dimension of 3 mm × 4
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6 mm × 45 mm for Young's modulus (E) measurement. The density of SCSZ and YSZ sintered
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9 were measured by Archimedes' principle.

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11 The Young's modulus in the temperature range between room temperature to 950 °C was measured
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13 by the impulse excitation technique [22]. The impulse exciting part of the device was set in an oven.
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15 The specimen, which had been dried in the oven at 60 °C for 24 h to remove any possible moisture at
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17 the surfaces, was hung by two thin platinum wires at the nodal points [22] in the oven. A ceramic pipe
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19 was fixed over the center of the specimen, and it extended outside the top of the oven. A steel ball can
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21 drop through the pipe onto the top center of the specimen to activate the impulse. There was another
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23 ceramic pipe located at the bottom center of the specimen, and it extended outside the bottom of the
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25 oven. The vibration of the impulse excitation was conducted by the pipe to the receiving part outside,
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27 which was a Grindo-Sonic Mk5 Industrial frequency analyzer (Lemmens, Belgium). During the
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29 measurement, the oven temperature increased from room temperature to 950 °C with a ramp rate of
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31 5 °C/min. The temperature was hold at every 50 °C for several minutes until it stayed constant, and
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33 then a steel ball was dropped through the upper pipe. The frequency analyzer collected the vibration
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35 and calculated the Young's modulus. Five measurements were carried out at each temperature point.
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47 PANalytical X'Pert Pro MPD Diffractometer (PANalytical B.V., Almelo, Netherlands) with a
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49 copper anode X-ray source with Bregg-Brentano geometry equipped with a high temperature stage
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51 (XRK 900 Reaction chamber, Anton Paar GmbH, Graz, Austria) was used for the *in situ* X-ray
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53 diffraction. The sintered YSZ and SCSZ specimens were ground into powders using a mortar and pestle.
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55 The YSZ and SCSZ powders were respectively heated to 275 °C and 350 °C with heating rate of
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57 2 °C/min, and they were isothermally annealed at those temperatures for 59 h and for 44 h, respectively.
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4 The X-ray diffraction patterns were collected every 12 minutes. The collected X-ray data were refined
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6 and analyzed by the software HighScore Plus.
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9 The neutron diffraction of YSZ powder was carried out [at the POWGEN instrument at Spallation](#)
10 [Neutron Source](#) (Oak Ridge National Laboratory, Oak Ridge, TN, USA). The powder was heated to
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12 275 °C with a heating rate of 5 °C/min and annealed at this temperature for 26 h using 80wt%N₂-
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14 20wt%O₂ flowing gas as the environment. To perform this heating experiment, the YSZ powder was
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16 placed in a quartz cylindrical can with vents on both ends for free gas flow. Quartz wool fibers were
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18 packed on both ends of the quartz tube to keep the powder inside of the can. High-resolution diffraction
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20 patterns were sequentially acquired every 64 minutes. The diffraction patterns were analyzed by
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22 Rietveld refinements using General Structure Analysis System package (GSAS) [23] and the graphical
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34 The vibrational properties of YSZ raw powders were studied with a Renishaw InVia Raman
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36 microscope (Renishaw Inc., Gloucestershire, UK). The Raman microscope system comprises a laser
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38 (532 nm line of solid Si) to excite the sample, a single spectrograph fitted with holographic notch filters.
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40 Before collecting spectra, the spectrometer was calibrated with a standard silicon wafer using a Si band
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42 position at 520.3 cm⁻¹. The YSZ raw powders were mounted inside a heating/cooling stage (TS1500,
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44 Linkam, UK) with a water circulation cooling system. The sample was heated up to 275 °C at a rate of
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46 2 °C/min, and then the temperature were dwelled for 216 h with the collection of Raman spectrum was
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48 performed every 20 min. The average collection time for a single spectrum was 300 s.
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61 **3 Results and Discussion**

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The SCSZ and YSZ ceramics were sintered to almost full density ($\rho = 5.679 \pm 0.003 \text{ g/cm}^3$ for SCSZ and $\rho = 5.883 \pm 0.019 \text{ g/cm}^3$ for YSZ). As a theoretical density of SCSZ and YSZ was calculated to be equal to 5.724 g/cm^3 and 5.914 g/cm^3 , the 0.79% and 0.54% porosity was calculated to exist in each material, respectively. A nominal 8mol% Y_2O_3 - ZrO_2 and 10mol% Sc_2O_3 -1mol% CeO_2 - ZrO_2 compositions were used for the calculation of the theoretical density without taking into account the possible impurities present in the powder. Young's modulus was measured to be $207.1 \pm 2.3 \text{ GPa}$ for SCSZ ceramics and $202.4 \pm 2.8 \text{ GPa}$ for YSZ ceramics at room temperature, which is in line with reported Young's modulus values [16,18]. The dependencies of Young's moduli of SCSZ and YSZ ceramics as a function of temperature are presented in Figure 1. The Young's moduli for both materials remain almost constant in RT–150 °C temperature range with non-significant softening of the lattice due to an input of thermal energy. However, in the range of ~200–400 °C the measured values of the Young's modulus become too low to be considered reliable with the measurement technique used in the current studies. At 400 °C the Young's moduli were measured to be $112 \pm 29 \text{ GPa}$ and $146 \pm 2 \text{ GPa}$ for SCSZ and YSZ, respectively, which is explained by a significant softening of the lattice. The further decrease of Young's modulus was measured in 400–500 °C temperature range, where the lowest measured value $E = 88 \pm 17 \text{ GPa}$ is reported at 500 °C for SCSZ, but the lowest measured $E = 128 \pm 1 \text{ GPa}$ at 600 °C for YSZ. Upon further increase of temperature, the Young's moduli of both ceramics keep increasing with $E = 162 \pm 2 \text{ GPa}$ for SCSZ ceramics and $E = 151 \pm 2 \text{ GPa}$ for YSZ ceramics measured at 950 °C.

A number of publications reported a strong damping of these ceramics in 200–400 °C temperature range, explaining the appearance of several loss moduli peaks in dynamic mechanical loading [25-27]. Such damping behavior is reported for both cubic and tetragonal YSZ [19,28], as well as appearance of

damping occurs in SCSZ [16]. In most publications, the oxygen vacancies hopping around the yttrium or scandium substitutional cations was proposed where the formation and reorientation of elastic dipoles ($V_O^{\bullet\bullet}-Y_{Zr}'$) between dopant cations and oxygen vacancies is a major mechanism contributing to the damping [20,29]. It was also found that for SCSZ, a slow cubic-to-rhombohedral (c-to- β) phase transition occurs in SCSZ in 200–400 °C temperature range, and therefore, it was hypothesized that such slow structural changes might also contribute to the appearance of the high loss modulus peaks in a specified temperature range. In order to study if similar structural changes from cubic phase to lower symmetry phase contributing to damping of YSZ ceramics at lower 200–400 °C might occur, the isothermal high temperature X-ray diffraction was performed on YSZ and SCSZ ceramics. In addition to *in situ* X-ray analysis, structural studies of YSZ were enhanced with isothermal high temperature neutron diffraction performed at the same temperature.

Figure 2 shows the profiles and intensity contour mappings of selected peaks in the X-ray diffraction patterns of SCSZ and YSZ ceramics during the isothermal annealing. As the range of the temperatures, where damping occurs, was different for SCSZ (175–400 °C) and for YSZ (200–400 °C) ceramics, the isothermal dwell temperatures were chosen 350 °C for SCSZ and 275 °C for YSZ in order to perform the isothermal annealing in the temperature range where damping is well pronounced.

The characteristic reflections (111) and (200), using pseudo-fluorite lattice, of the SCSZ at the beginning of the annealing (dwell time = 0 hours) and at the end of the isothermal heating (dwell time = 44 hours) are shown in Figure 2A. Similarly to the results reported earlier [8], the pristine pure cubic phase turned to be the mixture of cubic and β phases after 44 h isothermal annealing at 350 °C. The contour plot of (111) and (200) peaks demonstrate the phase evolution in SCSZ ceramics (Figure 2B–C). At the very beginning, only cubic phase could be found by XRD, and then the initiation of the

c-to- β phase transition was detected after 4 h dwell. Once the β -rhombohedral phase appears, the intensity of β phase peaks progressively increased with time, accompanied by the decrease of the cubic phase. From the Rietveld refinement, it is possible to calculate the weight fraction patterns of the phases (Figure 3). The dashed lines are placed at the beginning of the annealing because in this case the XRD was not sensitive to < 5 wt% phase fraction, and so it was not possible to determine the fraction of the β -rhombohedral phase. After 44 hours of dwell time at 350 °C, the SCSZ consisted of 56.7 wt% of cubic phase and 43.3 wt% of β -rhombohedral phase. In our previous publication a complete transformation from cubic to rhombohedral phase was observed after only 12 hours annealing at 350–400 °C [8], it was also reported that the grain size plays an important role on the kinetics of the phase transition in SCSZ. As the grain size of the SCSZ ceramics used for this investigation was about 10 μm , the c-to- β phase transition was not completed even after 44 hours of isothermal annealing, and most likely two phases would coexist upon longer period of time during dwell at 350 °C.

X-ray diffraction analysis of YSZ did not reveal any phase changes in this temperature range and only cubic fluorite phase is reported to exist, as reflected by (111) and (220) diffraction peaks of YSZ (Figure 2D) and the contour plots as a function of dwell time (Figure 2E–F). There was no change detected in the lattice parameter of the YSZ ceramic upon dwelling at the isothermal temperature, where the lattice parameter a was calculated to be equal to 5.15032 ± 0.00004 Å and the unit cell volume was calculated to be equal to 136.616 ± 0.003 Å³. They remained constant for the whole duration of isothermal annealing.

While no change in the structure by XRD is reported for YSZ ceramics, significant changes in the lattice parameters were found both in cubic and in rhombohedral phases during annealing of SCSZ (Figure 3). As pure cubic phase of SCSZ started transforming to the rhombohedral phase after about

250 min of dwell time at 350 °C, the two phases were forced to coexist. During such dwell the lattice parameters of each phase were continuously changing. For the first 500 min of the dwelling time, the lattice parameter a of the cubic phase exhibit slight increase from 5.1071 Å at the beginning of dwell time to 5.1072 Å at 500 min of annealing. However, after that, the decrease of the lattice parameter began and it reached about 5.1068 Å value after 2640 min of the annealing. When β -rhombohedral peaks started to appear at about 270 min of annealing, a_R and c_R (i.e., in hexagonal coordinates) did not remain constant but changed significantly during high temperature annealing. When β phase began to appear, a_R had lower value of 3.5769 Å than those exhibited by the prototype cubic structure, but it increased to 3.5787 Å after 800 min of annealing and remained almost constant till the very end of isothermal dwell. In contrast, lattice parameter c_R was equal to 9.0185 Å when the β phase first appeared and it decreased to 9.0145 Å after the first 800 min of annealing. It kept decreasing up to the very end of isothermal dwell shown 9.0138 Å value at 2640 min of annealing. The estimated standard deviations were large at the beginning due to the rather small amount of β phase yielded. Though the error bars are fairly large, the trends in Figure 3 clearly demonstrate the subtle changes of SCSZ lattice described above.

In comparison with cubic phase with $1/2[110] = 3.611$ Å and $[111] = 8.846$ Å, which are equal to a_R and c_R in the rhombohedral coordinate system used for β phase, it is seen that there are discontinuities in the lattice parameters between prototype cubic and β -rhombohedral phase, and thus the c-to- β phase transition can be considered as a first order phase transition [30].

By using calculated lattice parameters both cubic and rhombohedral structures of SCSZ, the volumes of unit cells were calculated (Figure 3). The volume of the cubic phase naturally follows the trend of a_c lattice parameter, where it increase slightly from 133.20 Å³ to 133.21 Å³ upon first 250 min

of annealing, however, when β phase started to appear the volume of the cubic unit cell started slowly decreasing and became equal to $133.18 \text{ \AA}^3$ at the end of annealing. The volume of the rhombohedral unit cell, when the phase just appeared, had a value of $133.22 \text{ \AA}^3$, which was very close to the values of the cubic lattice, but it rapidly increased within next 1000 min of the dwell time to $133.32 \text{ \AA}^3$, and upon further annealing the volume of β phase unit cell was gradually decreasing until it became $133.30 \text{ \AA}^3$ after 2500 min of annealing (Figure 3). Thus the kinetics of c-to- β phase transition in SCSZ was clearly seen from isothermal X-ray diffraction.

Therefore, the reason for the damping occurring in SCSZ during measurement of the Young's modulus by impulse excitation technique at 250–400 °C temperature range could be the reported phase transition, which possibly requires significant structural preparations such as nucleation of β phase in the cubic matrix following with the further growth of the phase amount accompanied by the simultaneous growth of lattice parameters. As a phase transition existed in SCSZ, the question was raised if it would be possible that a similar, but much slower, unreported phase transition to lower symmetry occurred in cubic YSZ, which was not taken into account in the previous vastly reported research. Our results with X-ray diffraction on the study of the crystal structure of YSZ by isothermal annealing of the ceramics at 275 °C for 59 hours indicated that only cubic phase ($Fm\bar{3}m$) existed as only peaks corresponding to cubic lattice can be found in the diffraction patterns, and no (111), (200) or (400) peaks' splitting can be found to indicate a phase transition to a lower symmetric structure. However, as the crystal structure analysis was performed utilizing X-ray techniques that are not really sensitive to detect changes in the location or quantities of light elements, the displacement of oxygen atoms in fluorite structure would not be detected. Therefore, other techniques, such as neutron diffraction or micro-Raman spectroscopy might provide a better insight into YSZ structure upon

isothermal annealing and determine the positions and occupancy of the oxygen lattice sites.

To verify the stability of cubic structure, neutron diffraction was performed where diffraction patterns were collected upon annealing of YSZ powders at 275 °C for 24 hours. The typical diffraction pattern with a result of the Rietveld refinement is shown in Figure 4A. Among all fitting patterns at 275 °C, the fitting parameters wRp , Rp and reduced χ^2 were found to be in the range of 3.5%–4.2%, 7.1%–10.6% and 1.3–1.41, respectively. All strong peaks in YSZ neutron diffraction pattern corresponded well to $Fm\bar{3}m$ cubic structure, however a set of forbidden peaks has been also detected. The presence of these peaks indicates the loss of cubic symmetry and appearance of lower symmetry tetragonal structure with $P4_2/nmc$ space group. One of such peaks, (112), is shown in Figure 4A (insert).

The calculated values of lattice parameters, volume of the unit cell, the oxygen position as well as oxygen site occupancy in the tetragonal fluorite YSZ structure at heating up and during isothermal annealing at 275 °C for 1500 min are plotted in Figure 4B–E. Both lattice parameters and volume of the tetragonal unit cell increased linearly as the temperature increased from RT to 275 °C. The calculation of the thermal expansion of YSZ upon heating in this temperature range provided us with a value of coefficient of thermal expansion equal to $9.16 \times 10^{-6} \text{ }^\circ\text{C}^{-1}$, which coincides well with the values in the previous report [31].

However, unlike during the isothermal dwelling of SCSZ, both lattice parameters of the tetragonal YSZ remained constant at the 275 °C annealing, indicating no structural changes and high stability of tetragonal phase (Figure 4B), as expected from the literature. In the view of the atom positions inside the tetragonal lattice, the appearance of tetragonal distortion with very small c/a ratio means that not only $\text{Zr}^{4+}/\text{Y}^{3+}$ cation sublattice has a distortion, but oxygen ions are also distorted from their tetrahedral

sites in the anion's sublattice. At room temperature at the beginning of the heating, the $(0,0,z)$ coordinate of the oxygen at $4c$ site was equal to 0.0273 ± 0.0005 , upon heating to $275\text{ }^{\circ}\text{C}$ the z became equal to 0.0252 ± 0.0006 at $125\text{ }^{\circ}\text{C}$ and remained constant with a value of 0.0251 ± 0.0003 , which translate to $0.1298 \pm 0.0015\text{ \AA}$ oxygen displacement along c -axis in comparison with the position in the prototype cubic phase. The decrease of z value upon temperature rise from room temperature to $125\text{ }^{\circ}\text{C}$ indicates that the tetragonal distortion is decreasing and the structure becomes closer to the cubic symmetry at $125\text{ }^{\circ}\text{C}$, however upon further increase of temperature and for the duration of isothermal dwelling z value remains constant. For comparison, the estimation of displacement of $\text{Zr}^{4+}/\text{Y}^{3+}$ cations in tetragonal lattice, estimated as difference between c and a lattice parameters, gives $c - a = 0.0172\text{ \AA}$, which is almost an order of magnitude lower than the displacement of oxygen in the lattice. Therefore, most likely the tetragonal distortion in YSZ could be induced by the displacement of oxygen ions rather than the $\text{Zr}^{4+}/\text{Y}^{3+}$ cations. Since the c/a ratio is very small and close to 1, the YSZ tetragonal phase was tentatively identified as the t' structure [15,32].

The oxygen sites occupancy in $8\text{mol\% Y}_2\text{O}_3\text{-ZrO}_2$, as calculated by means of Rietveld refinement, is shown in Figure 4E as a function of temperature and dwell time. The oxygen occupancy did not exhibit significant deviation during the whole thermal treatment, with an average value of 0.94 ± 0.01 . This value is smaller than the theoretical oxygen vacancy of 0.965 for 8 mol% Y_2O_3 substitution in ZrO_2 lattice.

Thus the results of structural characterization of YSZ powder by neutron diffraction, while reveal an expected lattice expansion upon heating and significant oxygen displacement and lower occupancies of oxygen lattice sites, did not indicate any temperature induced phase transition in YSZ upon isothermal annealing.

Raman spectroscopy was also employed to detect any possible structural changes in YSZ ceramics, which might be responsible for the damping during elastic properties measurements by acoustic technique. The YSZ raw powder was heated to 275 °C and isothermally annealed for 216 h with selected spectra collected during such *in situ* measurements presented in Figure 5. As it was reported in a number of other publications [13,28], the YSZ spectrum consists of bands belonging to the cubic fluorite structure but one band at 465 cm⁻¹ indicates the tetragonal distortion [13], which is in a good agreement with the results obtained by neutron diffraction. As one can see from Figure 5, there are no significant changes in Raman spectra of YSZ for the whole 216 h duration of annealing and the structure remain stable. Two changes can be detected: one change is a decrease of intensity of 465 cm⁻¹ band when temperature started increasing from RT till 125 °C, and another change is the appearance of two weak bands at 950 cm⁻¹ and 1014 cm⁻¹ wave numbers upon annealing. The first change in the intensity of 465 cm⁻¹ band upon heating to 125 °C correlates well with data on oxygen displacement in YSZ anion sublattice obtained by neutron diffraction. Such changes indicate the decrease of tetragonal distortion. However, upon further dwell at 275 °C the *t'* phase remained unchanged and did not disappear with time upon dwelling. Two other bands at 950 cm⁻¹ and 1014 cm⁻¹ might correspond to the bands due to impurities segregation in the material during annealing. A separate study might be needed to determine the nature and origin of these bands.

4 Conclusions

As a result of the performed research, a transition from cubic phase to β -rhombohedral phase was confirmed to exist in SCSZ ceramics upon isothermal annealing at 350 °C. The appearance of the phase

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4 transition could be one of the reasons for the damping occurring in the materials during Young's
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6 modulus measurements by impulse excitation technique. On the opposite, no phase transition was
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8 detected to occur during isothermal annealing of YSZ ceramics at 275 °C. The oxygen ion shifting in
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10 YSZ at 125 °C might have relation to the oxygen-cation bonding, oxygen vacancy hopping and the
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12 dynamic damping behavior, but lack of more direct evidence. Thereby some other mechanisms rather
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14 than structural transition might be responsible for damping in Young's modulus of YSZ ceramics at
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16 high temperatures.
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Figure1

Figure2

Figure3

Figure4

Figure5

Figure Captions

Figure 1 Temperature dependence of Young's moduli in SCSZ (A) and YSZ (B).

Figure 2 (111) and (220) peak profiles by XRD ($K_{\alpha 2}$ stripped) of SCSZ (A) and YSZ (D) before and after annealing; intensity contour vs. annealing time mapping of SCSZ (B–C) and YSZ (E–F). Suffix C: cubic phase; suffix R: rhombohedral phase. Indexes are in pseudo-fluorite coordinates.

Figure 3 Weight percentages, lattice parameters and unit cell volumes (pseudo-fluorite) of cubic and rhombohedral phases in SCSZ during annealing at 350 °C by X-ray diffraction.

Figure 4 Neutron diffraction pattern of YSZ at 275°C annealing (A); and the lattice parameters (B), the volume of tetragonal unit cell (C), the coordinates of oxygen ions (D) and the occupancy of oxygen ions (E) of YSZ during temperature ramp and dwelling at 275 °C calculated by the refinement of neutron diffraction patterns.

Figure 5 Raman spectra of YSZ at the temperature ramp from RT to 275 °C and further dwelling for up to 216 h.