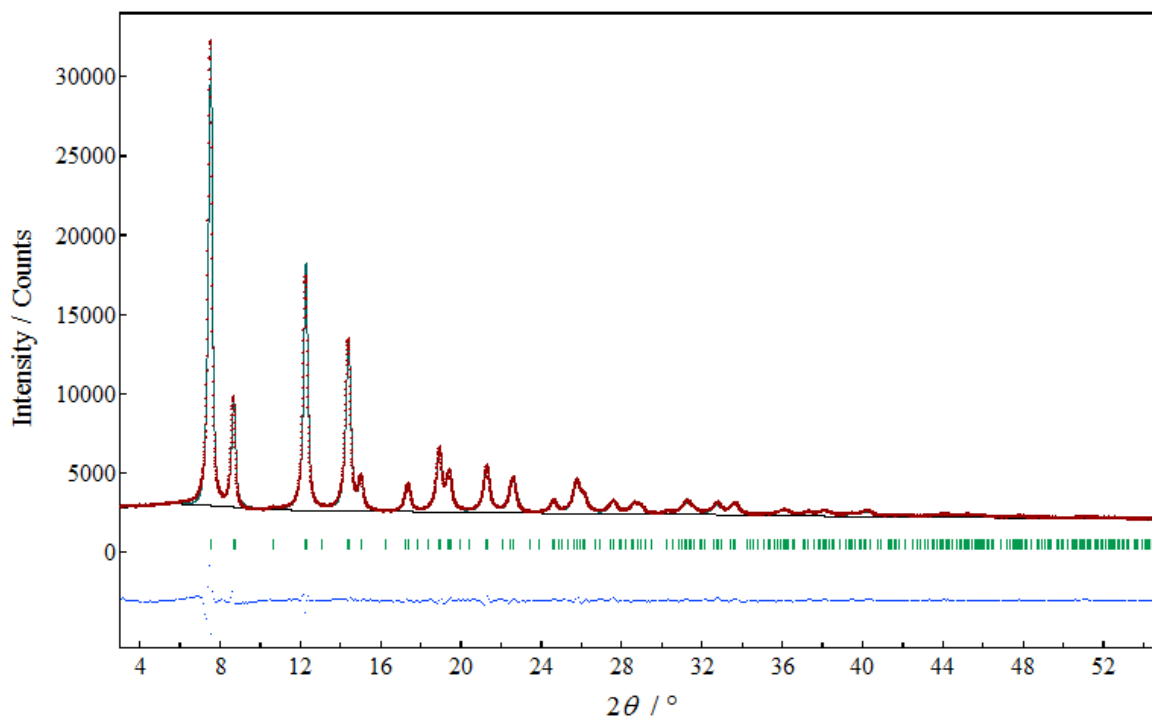## Supporting Information A:

**Photo S1.** Photo of the formation of the polymeric gel obtained by heating at about 140 ^{o}C.

Supporting Information B:

Fig. S1. Rietveld pattern of the synchrotron X-ray powder diffraction data of the nano-crystalline, compositionally homogeneous $Ce_{0.5}Zr_{0.5}O_2$ taken at 303 K (BL02B2 of SPring-8). This analysis indicates that the sample consists of a single phase of t'-form.

Supporting Information C:
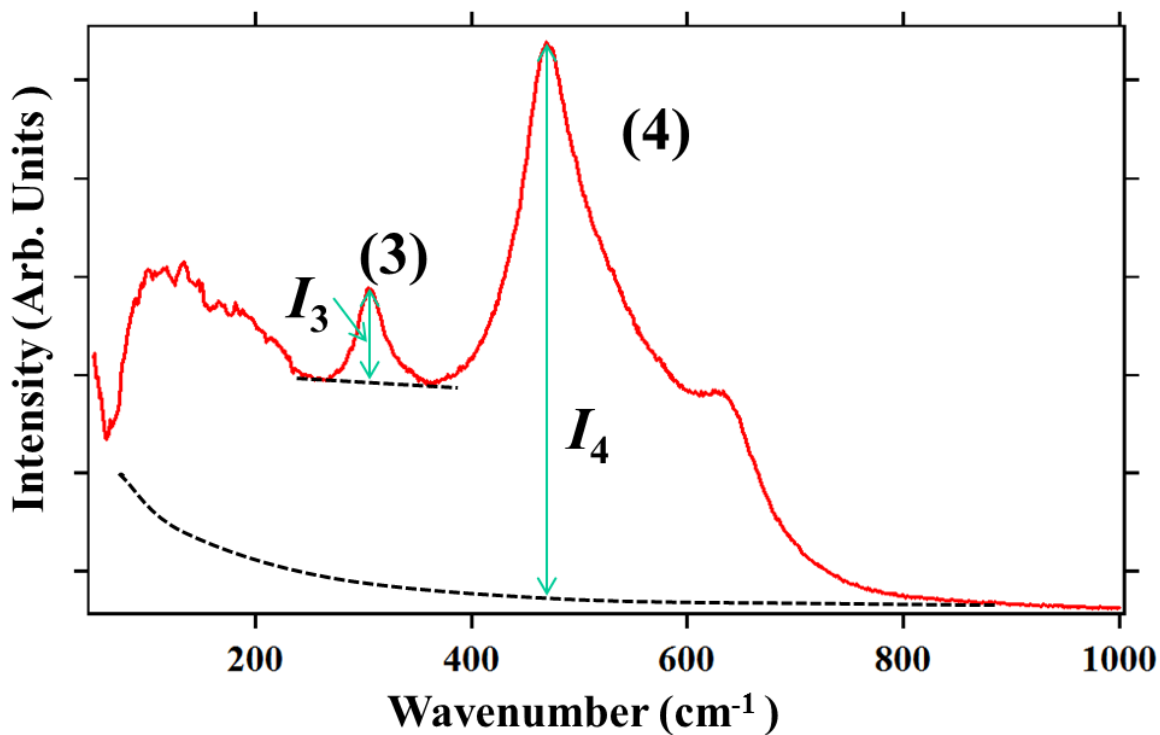
Fig. S2. Raman spectrum of nano-crystalline, compositionally homogeneous ceria-zirconia $Ce_{0.5}Zr_{0.5}O_2$. The Raman band (3) in Fig. S2 is characteristic of tetragonal symmetry, which is forbidden for the cubic symmetry. The normalized Raman intensity of the 3rd band was estimated to be $I_3/I_4 = 0.17$.

Supporting Information D:
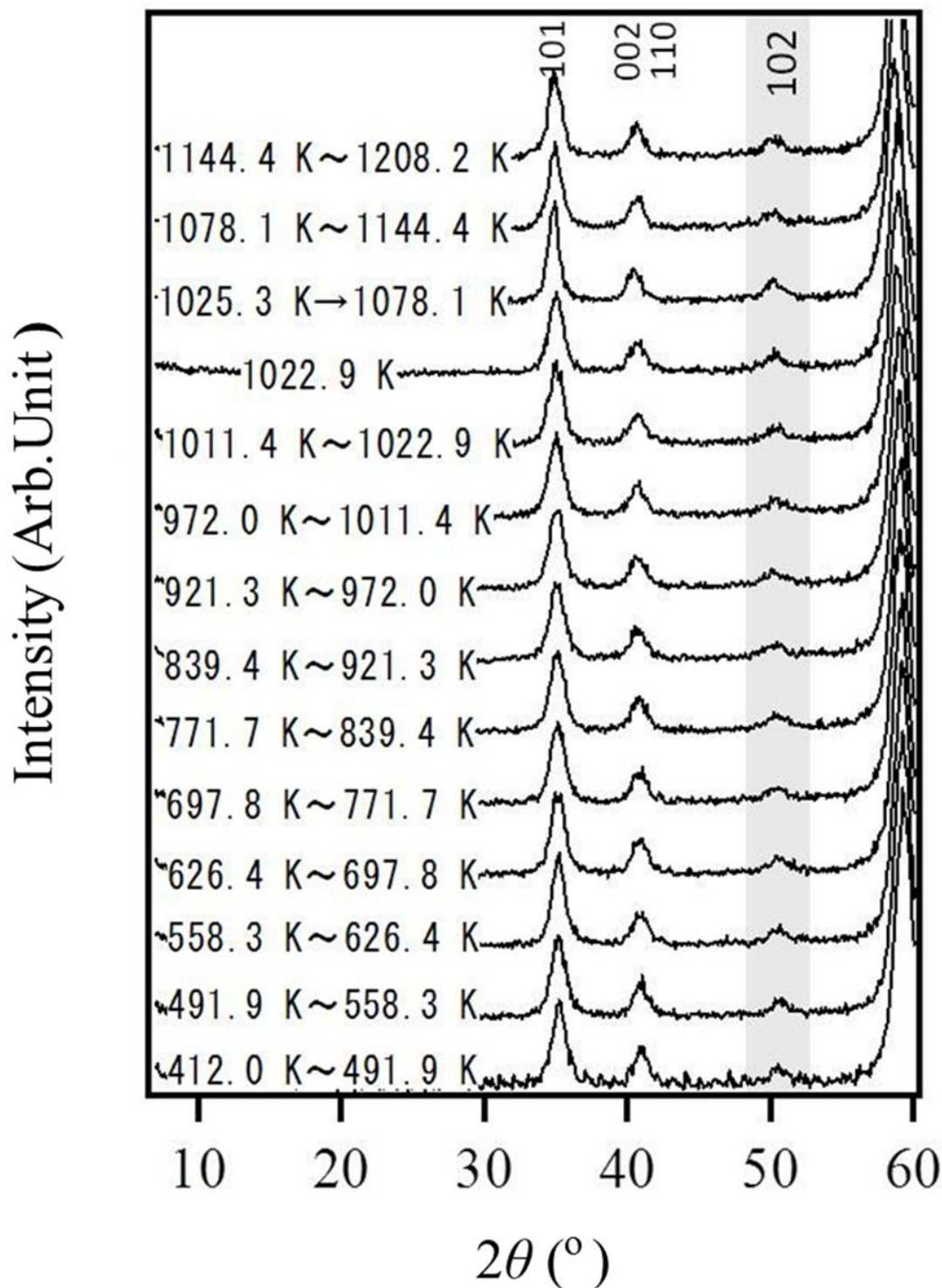
Fig. S3. Neutron-powder-diffraction profiles of the nano-crystalline, compositionally homogeneous $Ce_{0.5}Zr_{0.5}O_2$ measured *in situ* during heating or at a constant temperature in air, which indicates the existence of 102_{tet} reflection and no phase separation in the whole temperature range up to 1208 K. The nano-crystalline, compositionally homogeneous $Ce_{0.5}Zr_{0.5}O_2$ consists of a single phase of t'-form.

Supporting Information E:
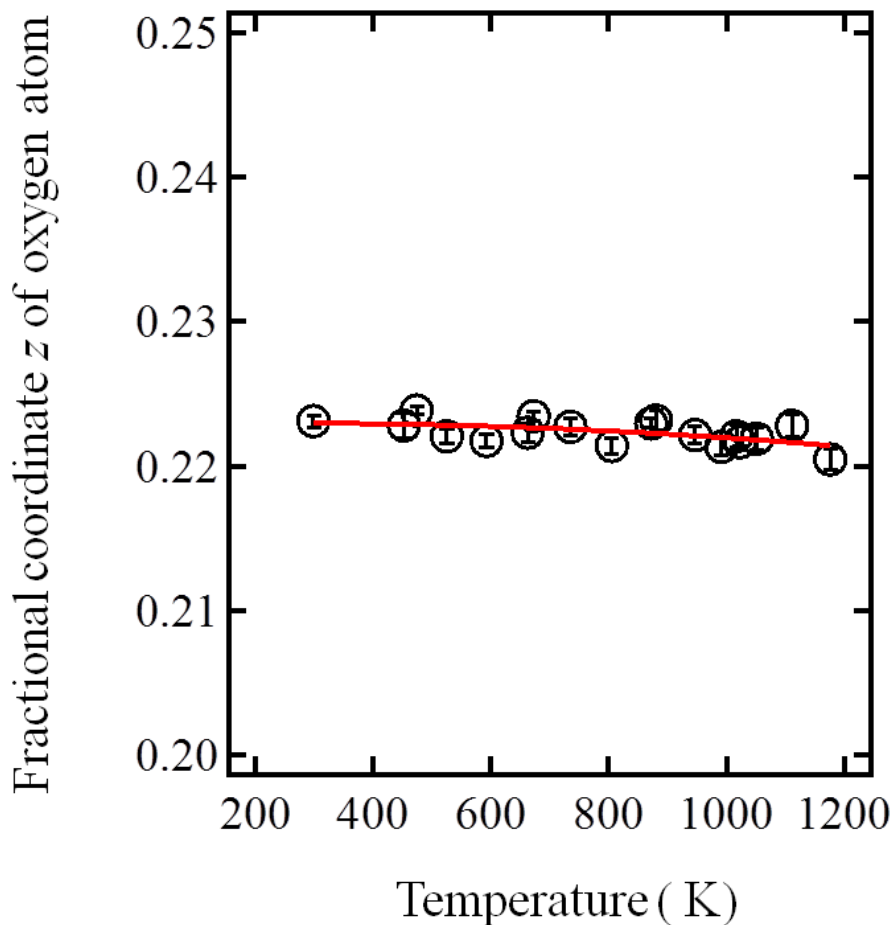
Figure S4. Temperature dependence of the fractional coordinate z of oxygen atom in the nano-crystalline, compositionally homogeneous $Ce_{0.5}Zr_{0.5}O_2$ solid solution. Error bar for each data is the estimated standard deviation in the Rietveld analysis. The solid line was obtained by the least-square means fit by a polynomial.

End of the supporting information.