Crystallite Size Estimation: Two techniques were used to estimate the average crystallite size: Rietveld refinement and Scherrer analysis. Qualitative trends in the average crystallite sizes were the same for both techniques. The average crystallite sizes determined by the Rietveld method were, however, systematically larger than those determined by Scherrer analysis. This behavior is expected because estimation using the Scherrer equation does not take into account strain explicitly.

Scherrer analysis is based on fitting the full with at half maximum of a given peak to the Scherrer equation,

\[ D = \frac{K\lambda}{\beta \cos \theta} \]

where \( K \) is the shape factor (0.9) and \( \lambda \) is the wavelength of the incident radiation, \( \beta \) is the full width of the peak at half maximum (in radians) and \( \theta \) is the Bragg angle (in radians).
**Figure S1.** Average crystallite size as estimated by the Scherrer equation.
**Figure S2.** Raman spectra of cerium oxide and cerium-zirconium oxide hollow spheres after various thermal treatments. Only signal from cerium oxide is observed (vertical dotted line at ~461 cm\(^{-1}\)).
Figure S3. Sample of the X-ray photoelectron spectra used for quantification of near-surface composition.
Figure S4. X-ray diffraction patterns as a function of temperature for $\text{Ce}_{1-x}\text{Zr}_x\text{O}_2$ samples, $x = 0.03$ (left), $x = 0.07$ (right). * represent peaks due to CeO$_2$ and # represent peaks due to the Pt stage.
Figure S5. Nitrogen adsorption isotherms of cerium oxide and cerium-zirconium oxide hollow spheres as synthesized and after various thermal treatments.