Ab initio and experimental oxygen ion conductivities in Sm-Zr and Gd-Zr co-doped ceria

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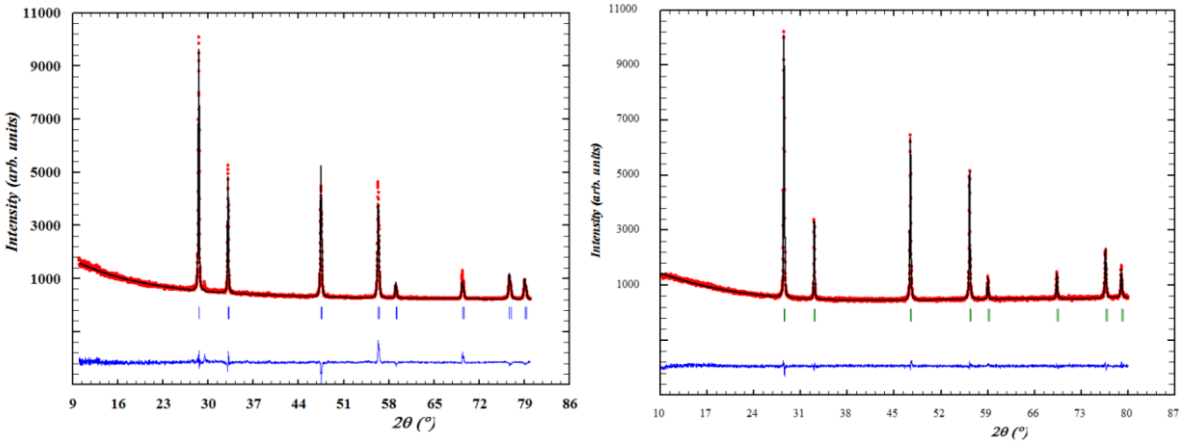

Figure S1: X-ray diffractogram and Rietveld refinement of $Ce_{0.887}Zr_{0.043}Sm_{0.070}O_{1.965}$ (left) and $Ce_{0.889}Zr_{0.006}Gd_{0.105}O_{1.9475}$ (right).[i]

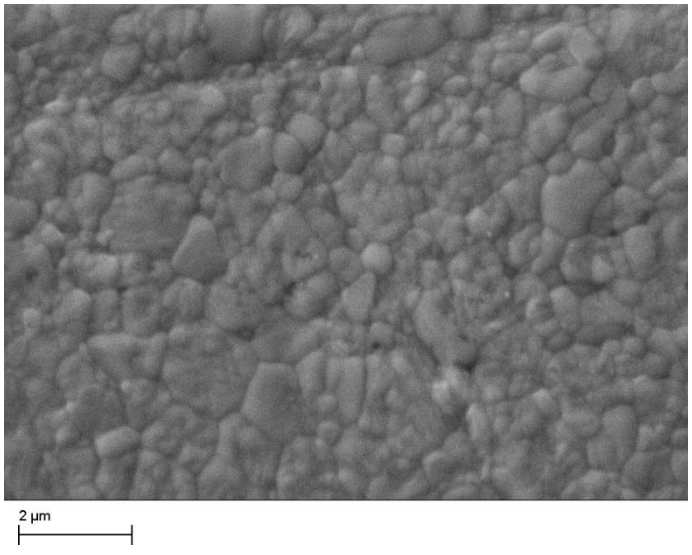

Figure S2: Scanning electron microscope measurement of $Ce_{0.88}Zr_{0.05}Sm_{0.07}O_{1.965}$.

Exemplary impedance plots for $Ce_{0.88}Zr_{0.05}Sm_{0.07}O_{1.965}$ at 379 °C for the polycrystal and the agglomerate of single crystals. The specific impedance $Z\frac{A}{l}$ with sample area A, and thickness l is shown for comparability. Note the different scales of the axes in Figure S1 and Figure S2. In the case of the single agglomerate, the semicircle for the grain boundary is strongly downscaled due to the small number of grain boundaries.

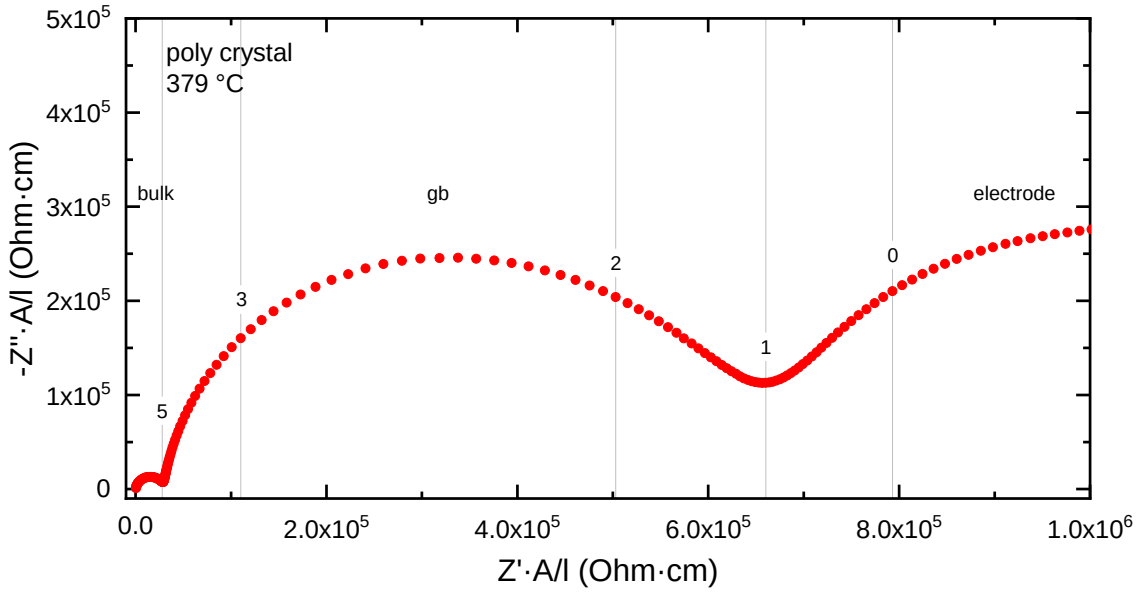

Figure S3: Impedance of $Ce_{0.88}Zr_{0.05}Sm_{0.07}O_{1.965}$ polycrystalline sample (grain size ~1 µm) at 379 °C. Geometry factors A and l are included in the values. Numbers in the graph state log_{10} of the respective frequency.

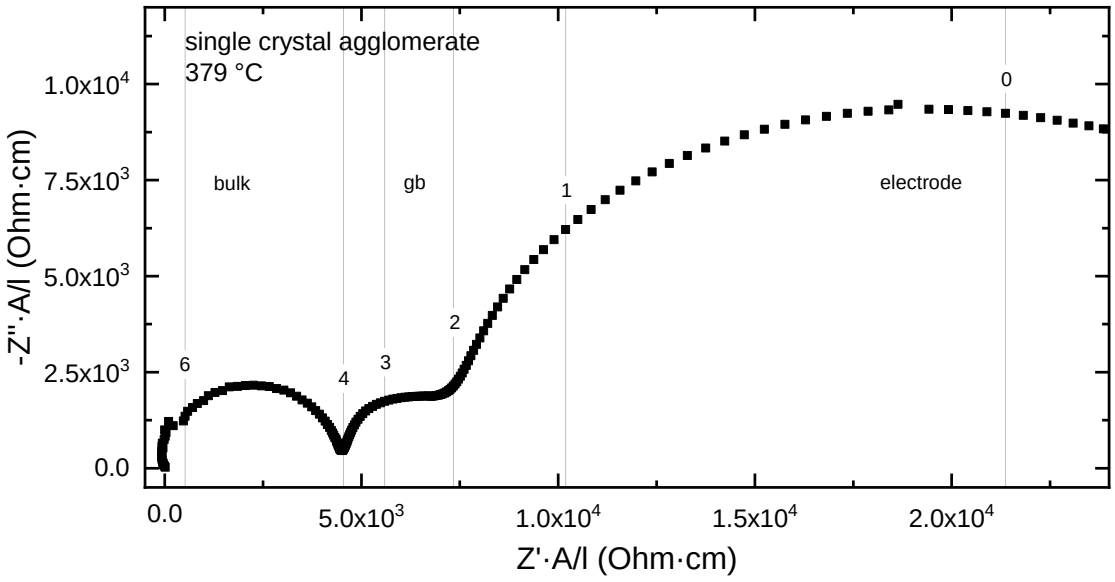

Figure S4: Impedance of $Ce_{0.88}Zr_{0.05}Sm_{0.07}O_{1.965}$ single crystal agglomerate sample (grain size ~500 µm) at 379 °C. Geometry factors A and l are included in the values. Numbers in the graph state log_{10} of the respective frequency.

Table S1: Migration energies for the specific edge configurations from density functional theory calculations in $Ce_{1-y}Zr_yO_2$.

edge	edge energy	
	$y = 0$	$y = 0.1$
Ce-Ce	0.466	0.493
Ce-Gd	0.577	0.638
Ce-Sm	0.631	0.699
Ce-Zr	0.251	0.301
Gd-Gd	0.901	0.951
Gd-Zr	0.392	0.430
Sm-Sm	1.031	1.091
Sm-Zr	0.427	0.474
Zr-Zr	0.139	0.163

Table S2: Pair interaction energies from density functional theory calculations in $Ce_{1-y}Zr_yO_2$. Interactions are labeled sequentially according to Figure 5 in the manuscript.

interaction	interaction energy	
	$y = 0$	$y = 0.1$
GdV1	-0.210	-0.227
GdV2	-0.060	-0.060
ZrV1	-0.600	-0.581
ZrV2	-0.050	0.014
SmV1	-0.150	-0.148
SmV2	-0.080	-0.060
VV1	0.848	0.895
VV2	0.291	0.305
VV3a	0.306	0.305
VV3b	0.358	0.421
VV4	0.268	0.272

[i] G. Ulbrich, Ph.D. Thesis, Technische Universität Berlin, Berlin, 2015.