

Supplemental Materials

Defect Accumulation in Swift Heavy Ion-Irradiated CeO₂ and ThO₂

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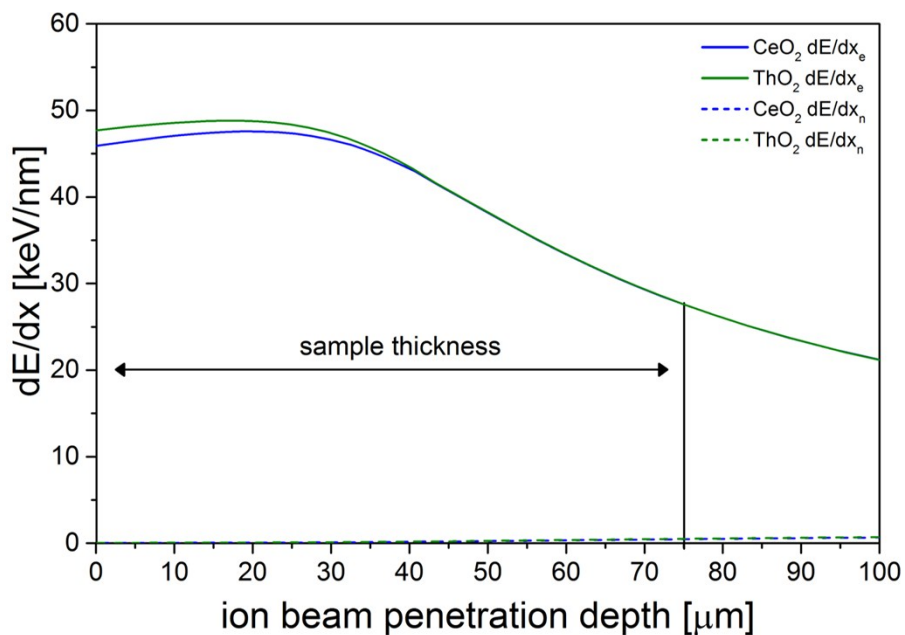
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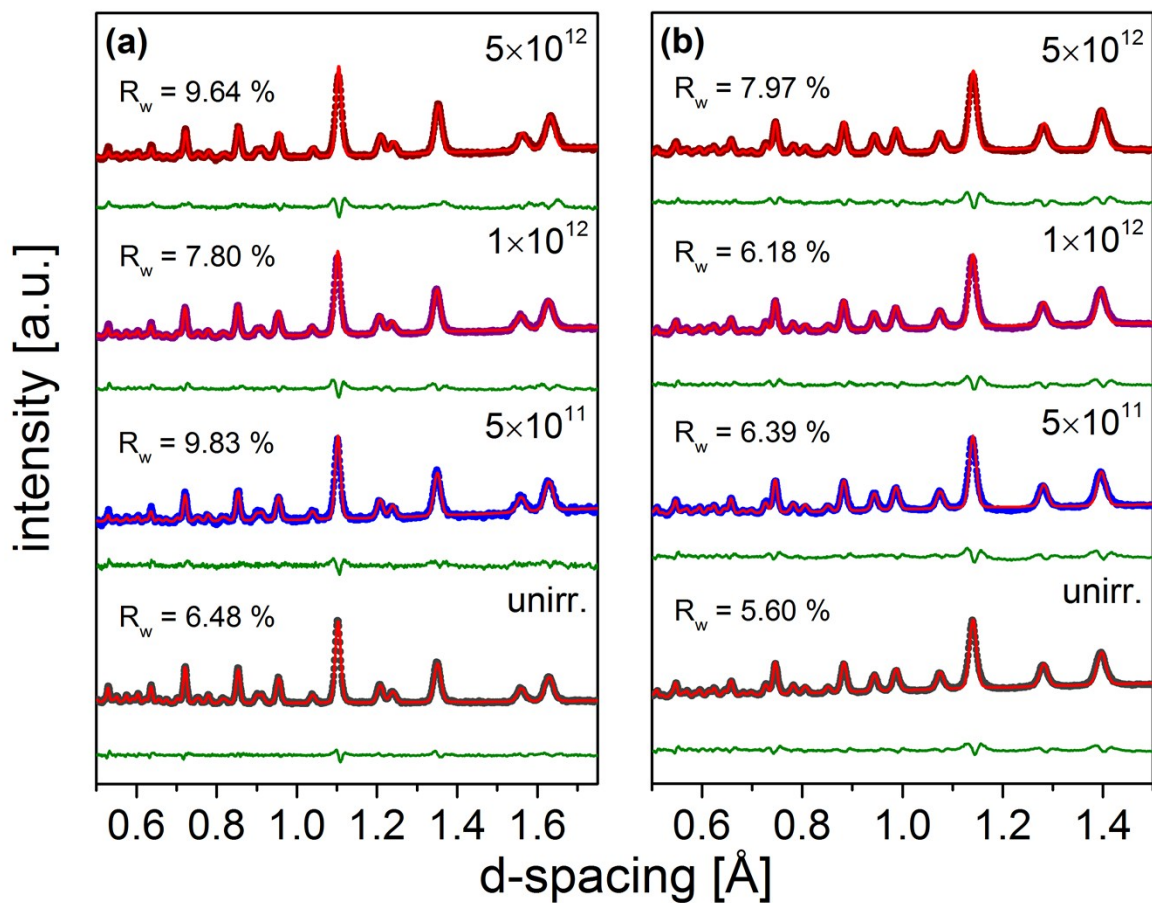
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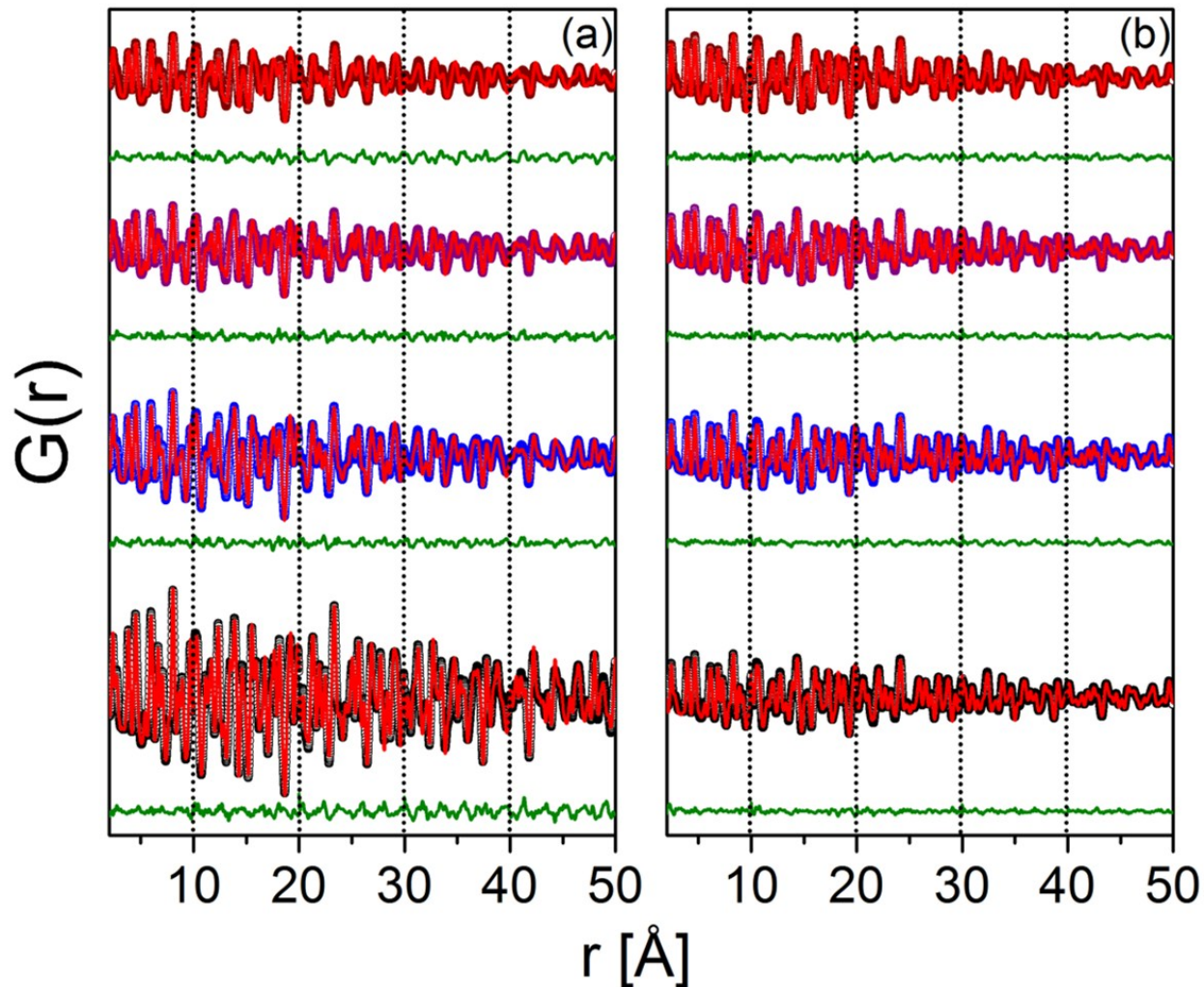
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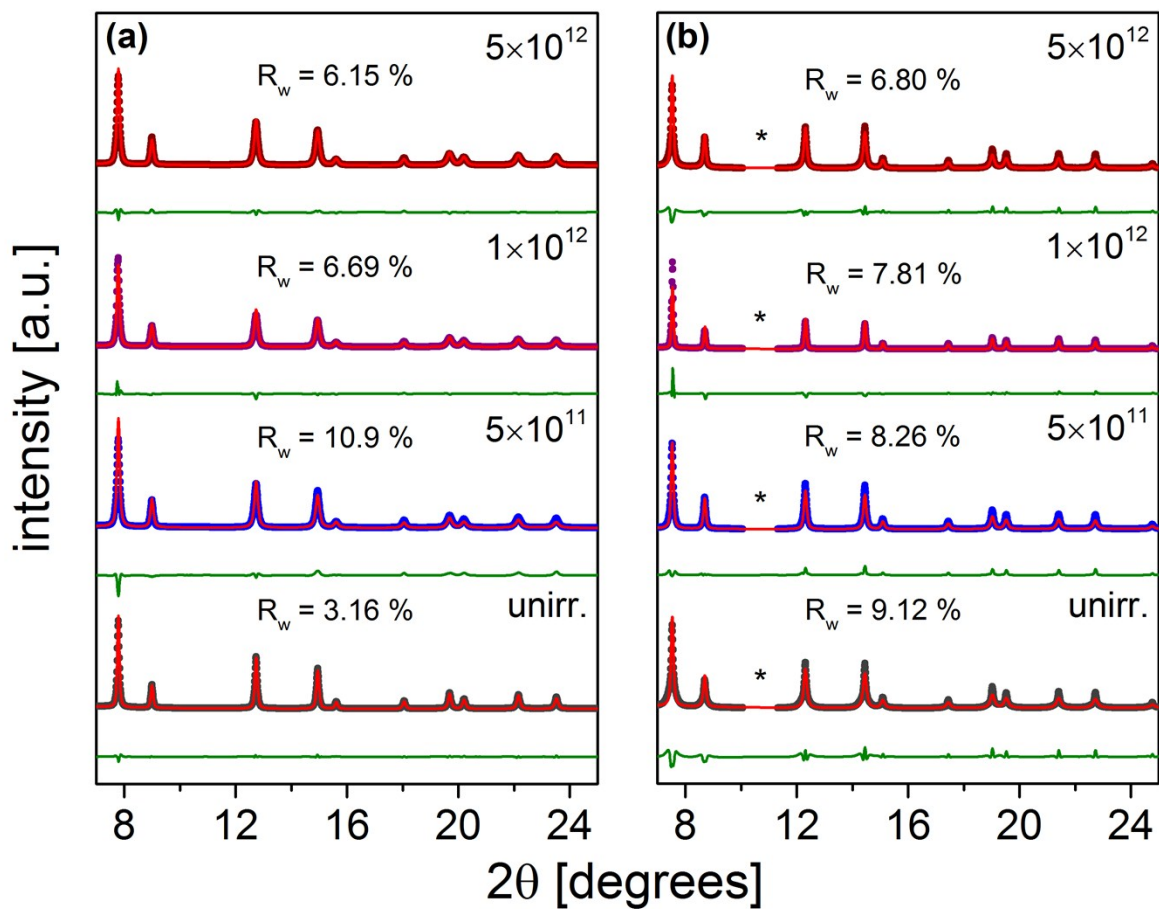
Supplemental Figure 1: Linear energy loss as a function of ion penetration depth in CeO₂ and ThO₂ calculated by SRIM-2010¹. The average electronic energy loss (*solid lines*) across the sample thickness (~75 μm) is 41 keV/nm and 42 keV/nm for CeO₂ and ThO₂, respectively. The corresponding nuclear energy loss (*dashed lines*) is negligible.



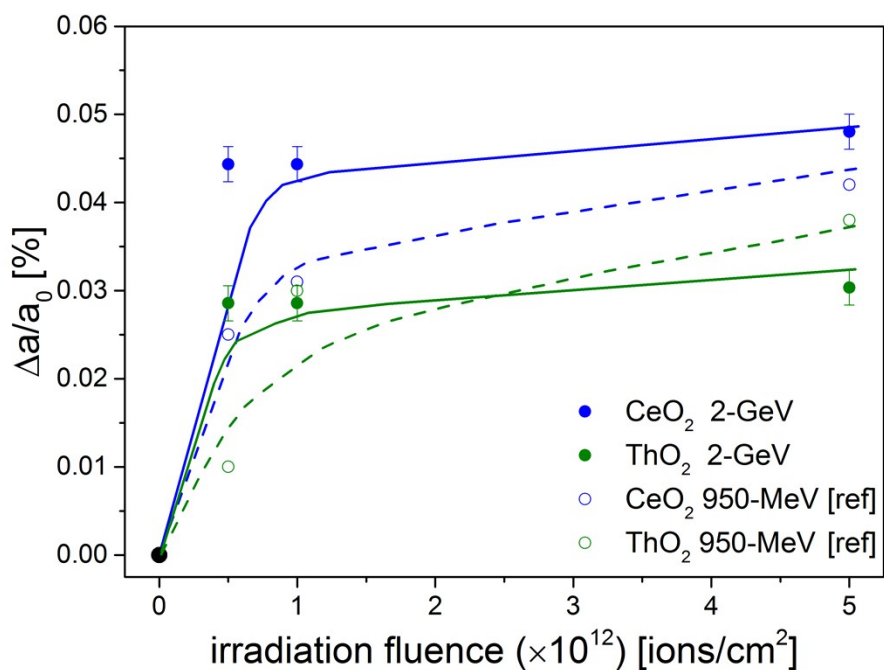
Supplemental Figure 2: Refined neutron diffraction patterns for all (a) CeO₂ (b) ThO₂ samples. The measured data, fitted profiles, and difference curves are represented by the colored circles, solid red lines, and solid green lines, respectively. Only the diffraction patterns from the $\theta = 66^\circ$ detector are shown for brevity. The fluence values are provided above each diffraction pattern.



Supplemental Figure 3: Refined pair distribution functions (PDFs) for all (a) CeO₂ and (b) ThO₂ samples. The measured PDFs, fitted fluorite structure models, and difference curves are represented by the colored circles, solid red lines, and solid green lines, respectively. The boundaries for the boxcar fits are denoted by the vertical, dotted black lines. The fits are representative of the ~ 10 Å-wide boxcar fits. Note: the four PDFs for a given material are not normalized to the same intensity; therefore, larger difference curves in this figure do not necessarily imply a larger R_w value. For a comparison of R_w values, refer to Figure 4 in the text.



Supplemental Figure 4: Refined synchrotron X-ray diffraction (XRD) patterns for all samples. The measured data, fitted profiles, and difference curves are represented by the colored circles, solid red lines, and solid green lines, respectively. The XRD peaks are much sharper compared to the Bragg peaks in the neutron diffraction patterns. This enabled the determination of more accurate unit cell parameters.



Supplemental Figure 5: Relative change in unit cell parameter, $\Delta a/a_0$, as a function of irradiation fluence as determined by Rietveld refinement of the X-ray diffraction patterns. The unit cell parameters obtained from X-ray and neutron diffraction are in agreement within experimental uncertainty. The lines are guides to the eye. The data are compared to data published for CeO₂ and ThO₂ samples irradiated with 950 MeV Au ions ². Both data sets illustrate similar irradiation behavior, albeit with different damage accumulation rates.

Supplemental Table 1: Interatomic distances derived from the pair distribution functions. The values represent the mean first nearest-neighbor cation-oxygen (<C-O>), oxygen-oxygen (<O-O>), and cation-cation (<C-C>) distances.

sample	<C-O>	<O-O>	<C-C>
CeO ₂ unirradiated	2.344(1) Å	2.711(1) Å	3.831(1) Å
CeO ₂ 5×10 ¹¹	2.332(1) Å	2.694(2) Å	3.829(1) Å
CeO ₂ 1×10 ¹²	2.293(10) Å	2.716(5) Å	3.837(1) Å
CeO ₂ 5×10 ¹²	2.349(2) Å	2.717(2) Å	3.837(1) Å
ThO ₂ unirradiated	2.419(1) Å	2.822(1) Å	3.960(1) Å
ThO ₂ 5×10 ¹¹	2.423(1) Å	2.784(1) Å	3.960(1) Å
ThO ₂ 1×10 ¹²	2.421(1) Å	2.810(1) Å	3.961(1) Å
ThO ₂ 5×10 ¹²	2.425(1) Å	2.784(1) Å	3.960(1) Å

References

1. J. F. Ziegler, M. D. Ziegler and J. P. Biersack, *Nuclear Instruments & Methods in Physics Research Section B-Beam Interactions with Materials and Atoms*, 2010, **268**, 1818-1823.
2. C. L. Tracy, M. Lang, J. M. Pray, F. Zhang, D. Popov, C. Park, C. Trautmann, M. Bender, D. Severin, V. A. Skuratov and R. C. Ewing, *Nat Commun*, 2015, **6**, 6133.