Supplementary Information for:

Correlating Nanoscale Secondary Ion Mass Spectrometry and Atom Probe Tomography Analysis of Uranium Enrichment in Metallic Nuclear Fuel

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Appendix 1: Sample preparation for NanoSIMS

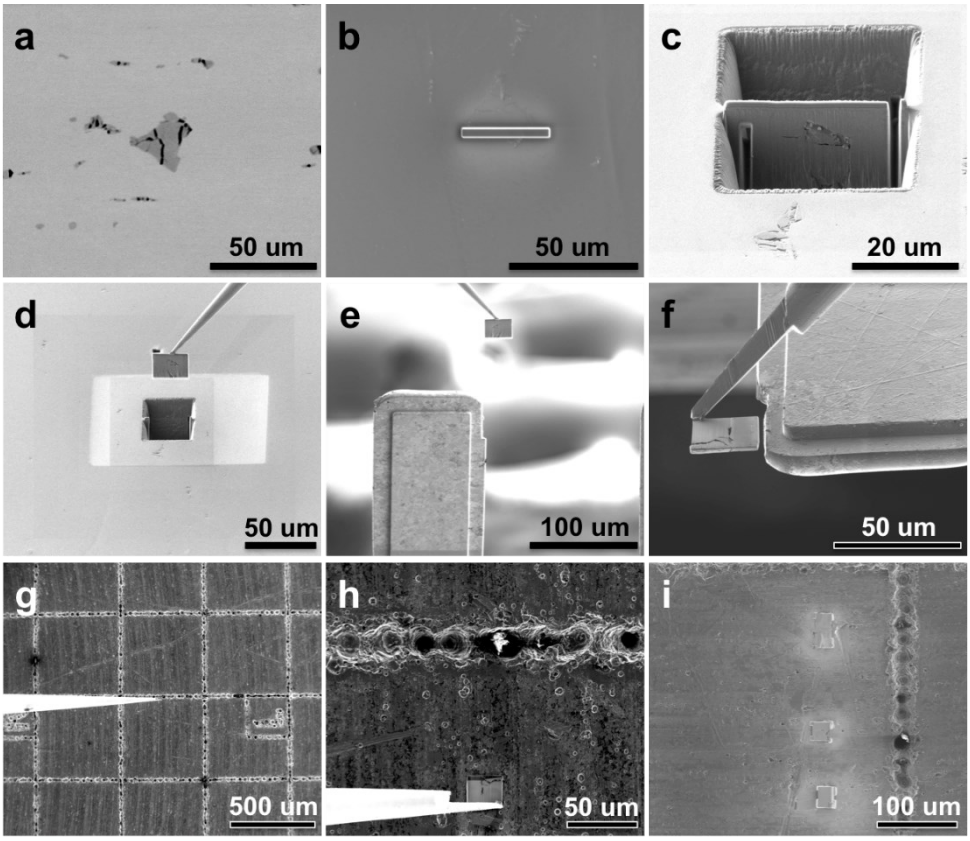

Figure S1: FIB based preparation of SIMS samples: (a) area of interest containing a UC inclusion is identified, (b) protective layer of C is deposited, (c) surrounding materials are moved and sample cut out, (d) sample is extracted, (e) sample is moved to and placed onto Cu half-grid, (f) sample is rotated and removed, (g-i) sample is moved to and deposited on a gridded wafer.

Appendix 2: Atom Probe Tomography Mass Spectra

Figure S2 shows an APT reconstruction including a UC/γ-UMo interface, also presented in the body of the manuscript (showing composition and enrichment across the interface). The 3D element distribution maps shown in Figure S2(a)-(d) show the distribution of U isotopes detected via APT (^{234,235,238}U). The full normalized mass spectra for γ -UMo and UC are shown in Figure S2 (e) and (g), respectively. The magnified view of U^{3+} isotopes in spectra are shown in Figure S2 (f) and (h) for γ -UMo and UC. ^{234}U, ^{235}U and ^{238}U are all detected via APT. To determine U isotopic composition via APT, the majority U species was used (U^{3+} elemental peak) [1]. The ranging criteria used for analysis is shown in Figure S3.

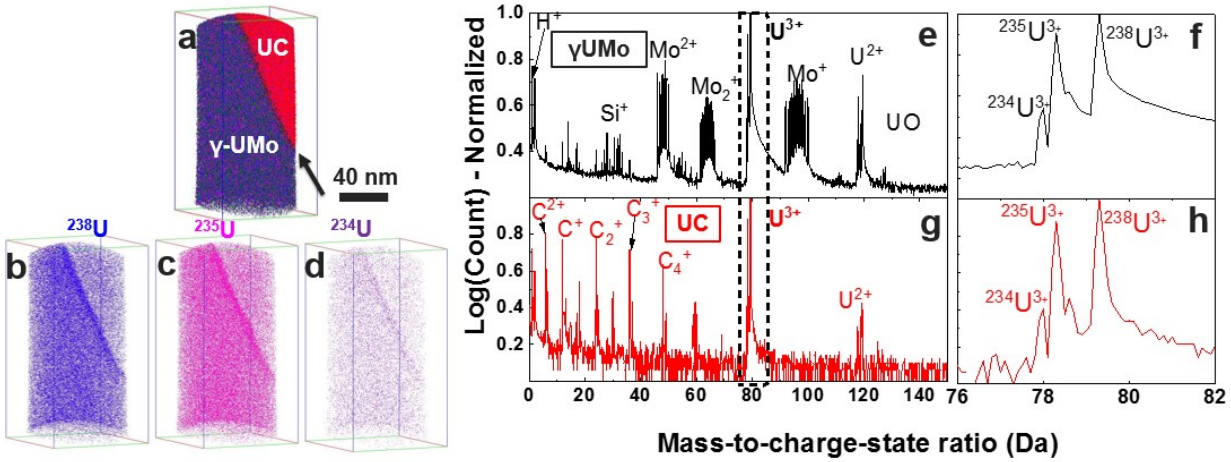

Figure S2. U isotope distribution across the γ-UMo/UC interface. (a) 3D element distribution maps of U, Mo, C and Si, (b) full mass spectra of the γ-UMo matrix phase, (c) mass spectra of the 76-84 Da range showing U^{3+} peaks (^{234,235,238}U) , (d) full mass spectrum for the UC phase, and (e) mass spectra of the 76-84 range for UC. 0.1 nm fixed bin width mass spectra are plotted above.

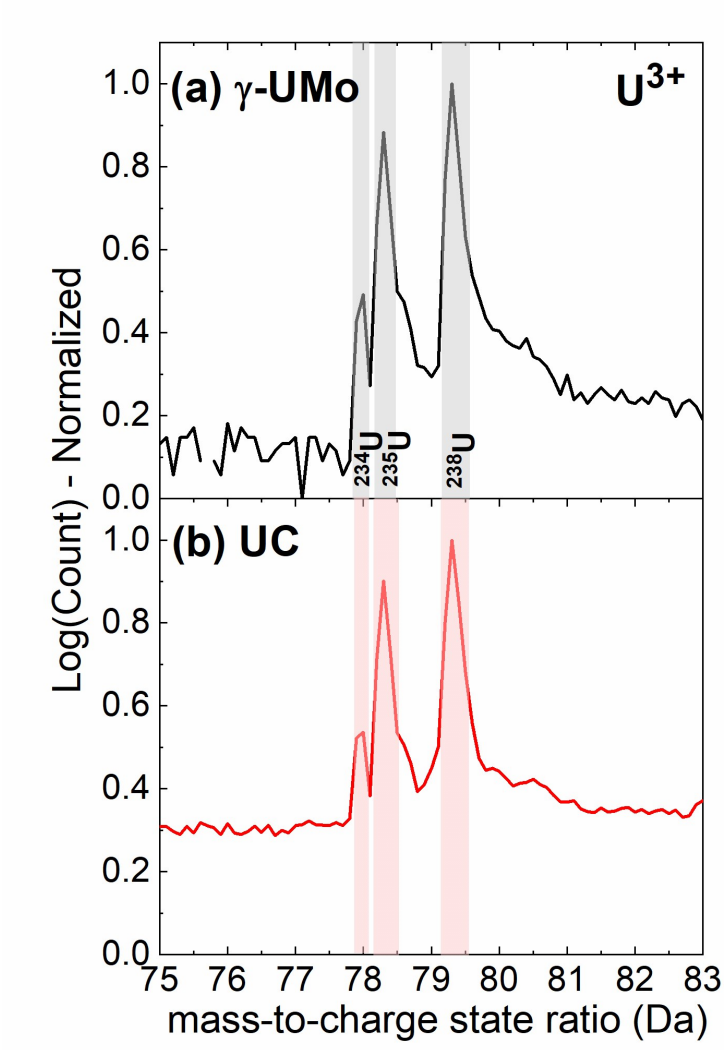

Figure S3. U isotope ranging criteria used in APT analysis of (a) γ-UMo and (b) UC phases. Each shaded box indicates the mass-to-charge state ratio ranged for each of the U isotopes.

Appendix 3: Atom Probe Tomography Data Sets and Analysis

For each data set analyzed and presented in the body of the manuscript, the number of ions, ranged ions, and ion counts for U isotopes are presented in Table S1. The average enrichment and standard deviation for each phase is also presented.

Table S1. Number of ions in the reconstructed volume, ranged ion counts, and ion counts for $^{234,235,238}U^{3+}$ for γ-UMo and UC APT data sets analyzed. Note that ^{235}U enrichment reported in this table was calculated considering ^{235}U and ^{238}U counts for comparison with NanoSIMS measurements.

Phase	Data Set	Spectrum Count	Spectrum Ranged Ion Count	Ion Count $^{238}U^{3+}$	Ion Count $^{235}U^{3+}$	Ion Count $^{234}U^{3+}$	at. % ^{235}U (enrichment)*
γ-UMo Matrix	1	3016097	2656152	1484398	367818.2	2166.598	19.86
	2	2769240	2430970	1372084	341265.9	2016.215	19.92
	3	4937493	4330250	2413802	607933.2	3874	20.12
	4	2178689	1916559	1153954	276289.7	1553.518	19.32
	5	7765072	7083146	4229451	1036536	5979.342	19.68
	6	16608694	8763052	1554659	390136	11501	20.06
	7	19,770,146	16,058,098	1.02E+07	2562693	17617.98	20.11
	8	7440022	6566129	4127063	962011.3	6706.446	18.90
	9	5040425+	4741575	2895118	718472	4449	19.88
				Average ± Standard Deviation			***19.76 ± 0.4***
UC	1	165366+	154712	83326	21177	151	20.26
	2	5076015	2680782	836032	208220	4583	19.94
	3	819968	629706	315840	80002.36	438.0024	20.21
				Average ± Standard Deviation			***20.14 ± 0.17***

+*These data sets are sub-sets of a single APT needle reconstruction that captured a UC/UMo interface (as seen in Figure S2). The UC data sets here refers to a sphere 30 nm by 30 nm by 30 nm in size. The enrichment was calculated from the mass spectrum analysis within this sphere.*

**235-U at. % calculated as follows:* $[235 - U^{3+} \,/\, (235 - U^{3+} + 238 - U^{3+})] \times 100$

Consideration of ^{234}U counts results in < 1 at. % change in calculated enrichment, and is not considered here for direct comparison with NanoSIMS data that calculates enrichment using ^{235}U and ^{238}U isotopes only.

References

[1] E. J. Kautz, D.E. Burkes, V.V. Joshi, C. Lavendar, A. Devaraj, Arun, "Nanoscale spatially resolved mapping of uranium enrichment " *Scientific Reports,* no. 9, 2019.