

Figure S1. Rietveld refinement of synchrotron powder XRD data for (a) U35T65 and (b) U71T29 samples. The black tick marks indicate the positions of the allowed diffraction maxima of the $I4_1/amd$ structure. The grey box indicates the range excluded from the fitting due to overlap with the nickel signal from the Inconel gasket.

Table S1. Unit cell parameters and R_{wp} (%) obtained by the Rietveld analysis of diffraction patterns for uranothorite samples with a zircon structure, collected at the APS beamline 6-IDB.

Sample	P (GPa)	a (Å)	c (Å)	R_{wp} (%)
U35T65	0.6(8)	7.062(4)	6.311(4)	4.57
	1.7(6)	7.052(4)	6.309(5)	3.66
	4.4(7)	7.003(6)	6.292(7)	4.34
	7.5(8)	6.951(7)	6.268(9)	4.08
	13(1)	6.85(1)	6.32(2)	4.38
U71T29	0(1)	7.026(3)	6.291(3)	5.27
	5.2(8)	6.949(5)	6.268(7)	4.03
	8.0(8)	6.917(6)	6.258(9)	4.20
	10.8(6)	6.885(7)	6.26(1)	4.39

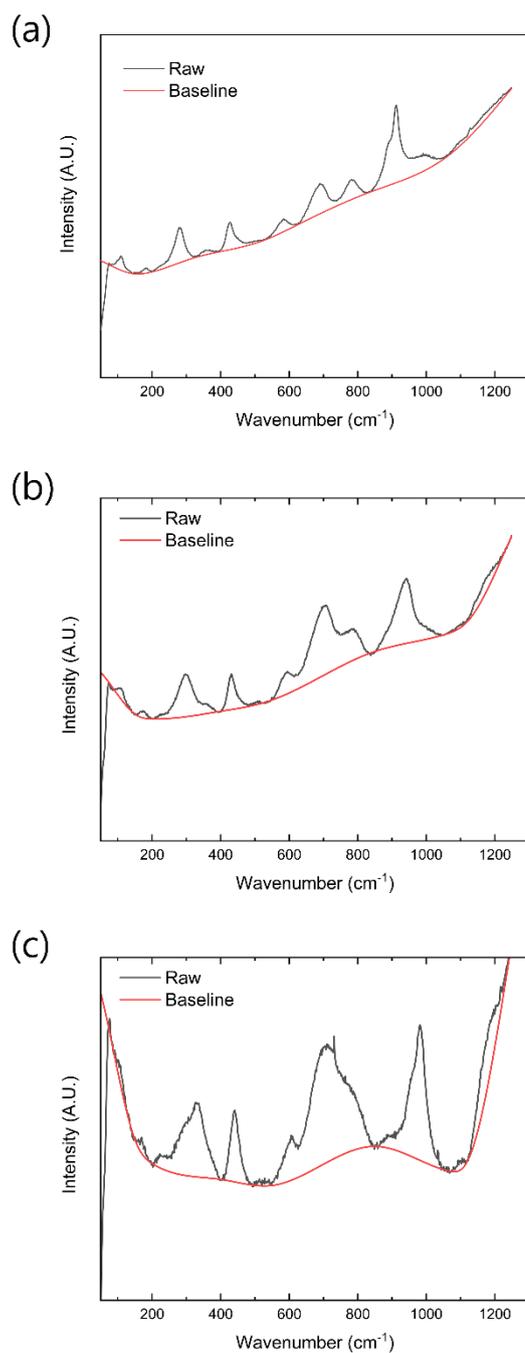


Figure S2. Representative Raman spectra of U35T65 at (a) 0, (b) 6.1 and (c) 15.2 GPa. The black lines represent the raw data, and the red lines the baseline used.

Peak Analysis

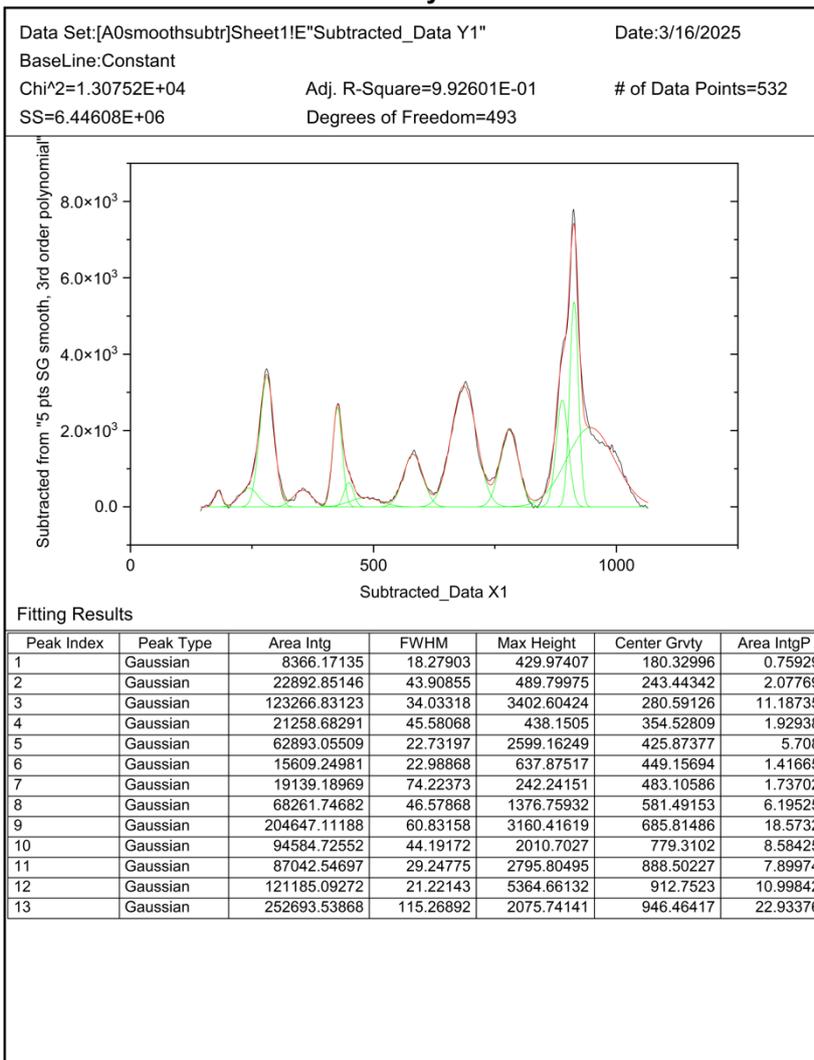


Figure S3. Representative fits of Raman spectra of U35T65 at 0 GPa.

Peak Analysis

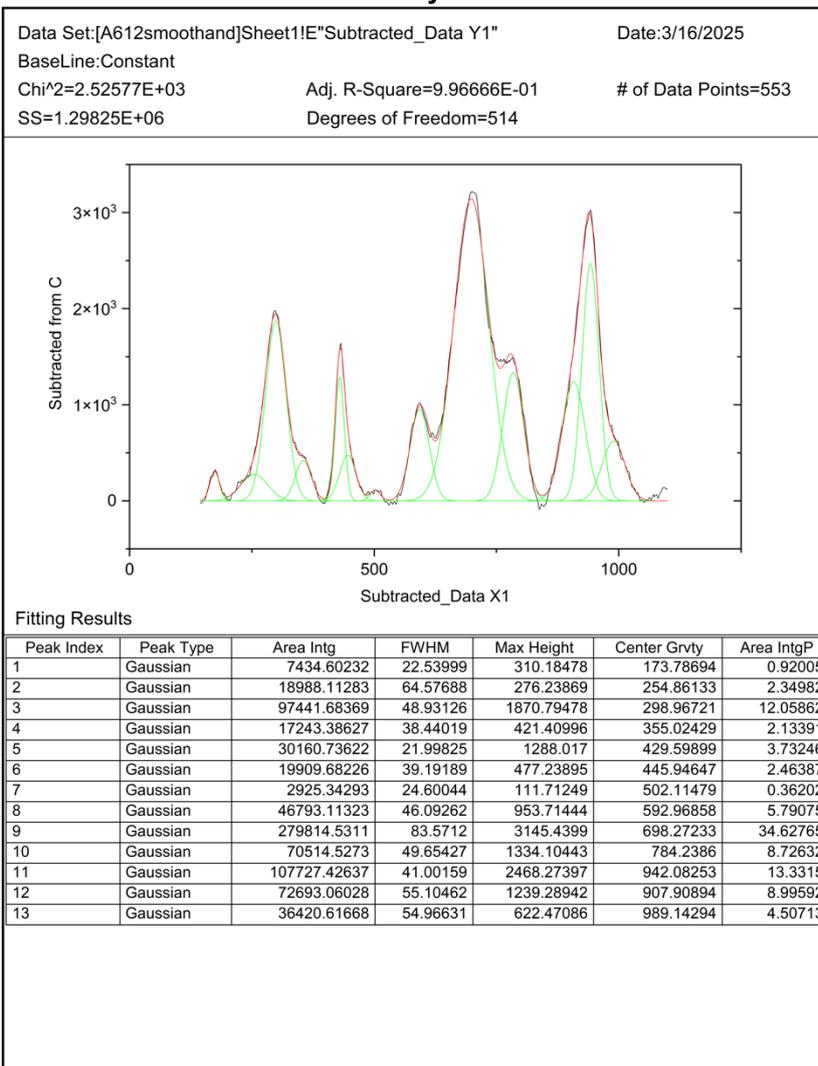
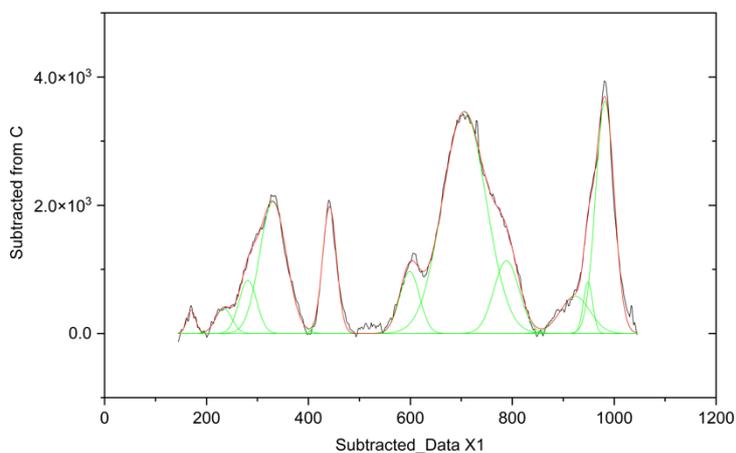


Figure S4. Representative fit of Raman spectra of U3T65 at 6.1 GPa.

Peak Analysis

Data Set:[A152all]Sheet1!E"Subtracted_Data Y1" Date:3/16/2025
 BaseLine:Constant
 Chi²=7.15719E+03 Adj. R-Square=9.93249E-01 # of Data Points=520
 SS=3.48555E+06 Degrees of Freedom=487



Fitting Results

Peak Index	Peak Type	Area Intg	FWHM	Max Height	Center Grvty	Area IntgP
1	Gaussian	7805.02073	20.24664	362.63796	170.10197	0.79976
2	Gaussian	15560.00843	37.31655	391.72037	233.64554	1.5944
3	Gaussian	36114.76253	40.96467	828.21502	281.22909	3.7006
4	Gaussian	134529.52117	61.5412	2053.61683	330.38512	13.78494
5	Gaussian	67249.03419	31.89892	1980.51362	441.36922	6.89086
6	Gaussian	47120.47144	45.72696	968.06634	598.69065	4.82833
7	Gaussian	371528.70059	100.9381	3457.84124	705.87186	38.0697
8	Gaussian	70087.85428	58.03805	1134.48238	788.63959	7.18174
9	Gaussian	42372.17346	68.61079	580.17993	923.44805	4.34178
10	Gaussian	18288.54412	21.32696	805.59737	948.60522	1.87399
11	Gaussian	165260.86628	42.88749	3620.9277	981.71822	16.93391

Figure S5. Representative fit of Raman spectra of U35T65 at 15.2 GPa.

Table S2. The second-order Birch-Murnaghan equations of states (EOS) parameters of coffinite, thorite, U35T65, and U71T29.

	EOS V_0 (Å ³)	K_0 (GPa)	pressure transmitting medium	notes
coffinite	306.2(4)	188(4)	16:3:1 methanol:ethanol:water mixture	Zhang et al. ¹
thorite	321.2(3)	142(4)	4:1 methanol:ethanol mixture	Strzelecki et al. ² using datapoints up to 6.2 GPa
U35T65	316.2(6)	170(9)		this study
U71T29	310.2(8)	215(11)		this study

Table S3. Changes in Gibbs energy and enthalpy with varying pressure. The thermal expansion coefficient values were adopted from Knyazev *et al's* work³.

Pressure (GPa)	$G_{f,P} - G_{f,0}$		$H_{f,P} - H_{f,0}$	
	$= \int_0^P (V_{huttonite} - V_{thorite}) dP'$		$= \int_0^P (V_{huttonite} - V_{thorite}) dP'$	$-\int_0^P (\alpha V_{huttonite} - \alpha V_{thorite}) T dP'$
0.1		-0.367074711		-3.75E-01
0.2		-0.731005959		-7.47E-01
0.3		-1.09180458		-1.12E+00
0.4		-1.449483611		-1.48E+00
0.5		-1.804053909		-1.84E+00
0.6		-2.155527112		-2.20E+00
0.7		-2.505255608		-2.56E+00
0.8		-2.853589071		-2.92E+00
0.9		-3.200535899		-3.27E+00
1		-3.54610304		-3.63E+00
1.1		-3.890296952		-3.98E+00
1.2		-4.233125607		-4.33E+00
1.3		-4.574595408		-4.68E+00
1.4		-4.914713412		-5.03E+00
1.5		-5.253486944		-5.37E+00
1.6		-5.590922044		-5.72E+00
1.7		-5.927025843		-6.06E+00
1.8		-6.261805692		-6.41E+00
1.9		-6.595267328		-6.75E+00
2		-6.927417517		-7.09E+00
2.1		-7.258263468		-7.43E+00
2.2		-7.587811235		-7.76E+00
2.3		-7.916067039		-8.10E+00
2.4		-8.243037925		-8.44E+00
2.5		-8.568729835		-8.77E+00
2.6		-8.8931487		-9.10E+00
2.7		-9.21630201		-9.43E+00
2.8		-9.538195165		-9.76E+00
2.9		-9.858833969		-1.01E+01
3		-10.17822517		-1.04E+01
3.1		-10.49637521		-1.07E+01
3.2		-10.81328938		-1.11E+01
3.3		-11.1289732		-1.14E+01
3.4		-11.44343384		-1.17E+01
3.5		-11.75667695		-1.20E+01
3.6		-12.06870808		-1.24E+01
3.7		-12.37953248		-1.27E+01
3.8		-12.68915693		-1.30E+01
3.9		-12.99758687		-1.33E+01
4		-13.30482772		-1.36E+01
4.1		-13.61088498		-1.39E+01
4.2		-13.91576491		-1.43E+01
4.3		-14.21947277		-1.46E+01
4.4		-14.52201383		-1.49E+01
4.5		-14.82339337		-1.52E+01

4.6	-15.12361758	-1.55E+01
4.7	-15.4226918	-1.58E+01
4.8	-15.72062096	-1.61E+01
4.9	-16.01741017	-1.64E+01
5	-16.3130646	-1.67E+01
5.1	-16.6075905	-1.70E+01
5.2	-16.9009929	-1.73E+01
5.3	-17.19327654	-1.76E+01
5.4	-17.48444624	-1.79E+01
5.5	-17.77450697	-1.82E+01
5.6	-18.06346466	-1.85E+01
5.7	-18.3513243	-1.88E+01
5.8	-18.63809073	-1.91E+01
5.9	-18.92376868	-1.94E+01
6	-19.20836268	-1.97E+01
6.1	-19.49187774	-2.00E+01
6.2	-19.77431938	-2.03E+01
6.3	-20.05569229	-2.06E+01
6.4	-20.33600116	-2.09E+01
6.5	-20.61525064	-2.11E+01
6.6	-20.89344528	-2.14E+01
6.7	-21.17058943	-2.17E+01
6.8	-21.44668838	-2.20E+01
6.9	-21.72174683	-2.23E+01
7	-21.99576929	-2.26E+01
7.1	-22.26876026	-2.28E+01
7.2	-22.54072419	-2.31E+01
7.3	-22.81166554	-2.34E+01
7.4	-23.08158859	-2.37E+01
7.5	-23.35049776	-2.40E+01
7.6	-23.61839792	-2.42E+01
7.7	-23.8852934	-2.45E+01
7.8	-24.1511885	-2.48E+01
7.9	-24.4160875	-2.51E+01
8	-24.67999465	-2.53E+01
8.1	-24.94291419	-2.56E+01
8.2	-25.2048503	-2.59E+01
8.3	-25.46580715	-2.61E+01
8.4	-25.72578868	-2.64E+01
8.5	-25.98479945	-2.67E+01
8.6	-26.24284368	-2.69E+01
8.7	-26.49992543	-2.72E+01
8.8	-26.75604875	-2.75E+01
8.9	-27.01121767	-2.77E+01
9	-27.26543618	-2.80E+01
9.1	-27.51870827	-2.83E+01
9.2	-27.77103788	-2.85E+01
9.3	-28.02242896	-2.88E+01
9.4	-28.2728854	-2.90E+01
9.5	-28.52241099	-2.93E+01
9.6	-28.77100951	-2.95E+01
9.7	-29.0186848	-2.98E+01
9.8	-29.2654407	-3.01E+01
9.9	-29.51128119	-3.03E+01

10	-29.75621014	-3.06E+01
10.1	-30.00023129	-3.08E+01
10.2	-30.24334836	-3.11E+01
10.3	-30.48556503	-3.13E+01
10.4	-30.72688499	-3.16E+01
10.5	-30.96731188	-3.18E+01
10.6	-31.20684932	-3.21E+01
10.7	-31.44550093	-3.23E+01
10.8	-31.68327029	-3.26E+01
10.9	-31.92016095	-3.28E+01
11	-32.15617647	-3.30E+01
11.1	-32.39132035	-3.33E+01
11.2	-32.62559603	-3.35E+01
11.3	-32.85900694	-3.38E+01
11.4	-33.09155656	-3.40E+01
11.5	-33.32324831	-3.42E+01
11.6	-33.55408561	-3.45E+01
11.7	-33.78407186	-3.47E+01
11.8	-34.01321044	-3.50E+01
11.9	-34.24150469	-3.52E+01
12	-34.46895796	-3.54E+01
12.1	-34.69557356	-3.57E+01
12.2	-34.92135477	-3.59E+01
12.3	-35.14630488	-3.61E+01
12.4	-35.37042712	-3.64E+01
12.5	-35.59372474	-3.66E+01
12.6	-35.81620094	-3.68E+01
12.7	-36.03785891	-3.71E+01
12.8	-36.25870182	-3.73E+01
12.9	-36.47873282	-3.75E+01
13	-36.69795505	-3.77E+01
13.1	-36.91637161	-3.80E+01
13.2	-37.13398558	-3.82E+01
13.3	-37.35080005	-3.84E+01
13.4	-37.56681806	-3.86E+01
13.5	-37.78204265	-3.89E+01
13.6	-37.99647681	-3.91E+01
13.7	-38.21012361	-3.93E+01
13.8	-38.42298625	-3.95E+01
13.9	-38.63506771	-3.98E+01
14	-38.84637093	-4.00E+01
14.1	-39.05689884	-4.02E+01
14.2	-39.26665435	-4.04E+01
14.3	-39.47564035	-4.06E+01
14.4	-39.68385971	-4.08E+01
14.5	-39.89131528	-4.11E+01
14.6	-40.0980099	-4.13E+01
14.7	-40.30394637	-4.15E+01
14.8	-40.50912751	-4.17E+01
14.9	-40.71355607	-4.19E+01
15	-40.91723483	-4.21E+01
15.1	-41.12016652	-4.23E+01
15.2	-41.32235386	-4.25E+01
15.3	-41.52379993	-4.28E+01

15.4	-41.72450756	-4.30E+01
15.5	-41.92447943	-4.32E+01
15.6	-42.1237182	-4.34E+01
15.7	-42.32222651	-4.36E+01
15.8	-42.52000699	-4.38E+01
15.9	-42.71706224	-4.40E+01
16	-42.91339486	-4.42E+01
16.1	-43.10900736	-4.44E+01
16.2	-43.30390224	-4.46E+01
16.3	-43.49808204	-4.48E+01
16.4	-43.69154992	-4.50E+01
16.5	-43.88430837	-4.52E+01
16.6	-44.0763599	-4.54E+01
16.7	-44.26770697	-4.56E+01
16.8	-44.45835205	-4.58E+01
16.9	-44.64829756	-4.60E+01
17	-44.83754594	-4.62E+01
17.1	-45.02609958	-4.64E+01
17.2	-45.21396102	-4.66E+01
17.3	-45.40113325	-4.68E+01
17.4	-45.58761862	-4.70E+01
17.5	-45.7734195	-4.72E+01
17.6	-45.9585382	-4.74E+01
17.7	-46.14297704	-4.76E+01
17.8	-46.32673832	-4.78E+01

References

1. Knyazev, A. V, Komshina, M. E. & Savushkin, I. A. Synthesis and high-temperature X-ray diffraction study of thorium orthosilicate. *Radiochemistry* 59, 225–228 (2017).