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Supporting information for article:

Investigation of local structure of molten ThF4-LiF and ThF4-LiF-BeF2 mixtures by high-temperature X-ray absorption spectroscopy and molecular-dynamics simulation

Jian Sun, Xiaojing Guo, Jing Zhou, Jianxing Dai, Sanzhao Song, Hongliang Bao, Jian Lin, Haisheng Yu, Shangming He, Feng Jiang, Dewu Long, Linjuan Zhang and Jian-Qiang Wang

1 Apparatus scheme for *in situ* high-temperature XAFS measurements

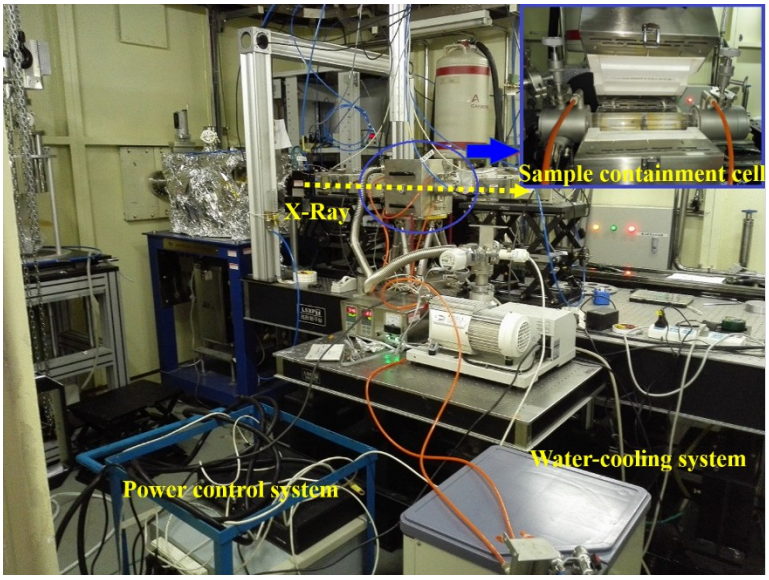

Figure S1 In-situ high-temperature XAFS measurement platform at the SSRF BL14W1 beamline.

S1. The phase of ThF_4-LiF-BeF_2 eutectic salt.

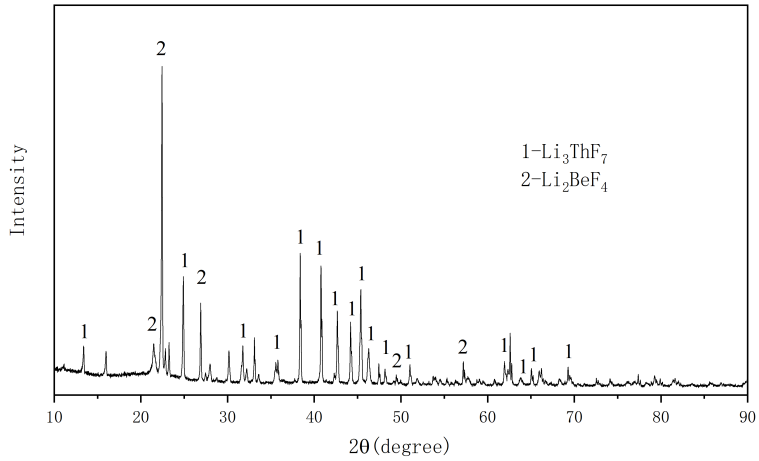

Figure S2 X-ray diffraction pattern of ThF_4-LiF-BeF_2 compounds, which mainly contains two phases (Li_3ThF_7 and Li_2BeF_4) when sample cooled into the room temperature.

S2. Comparison of EXAFS data before and after subtracting the double-electron excitation

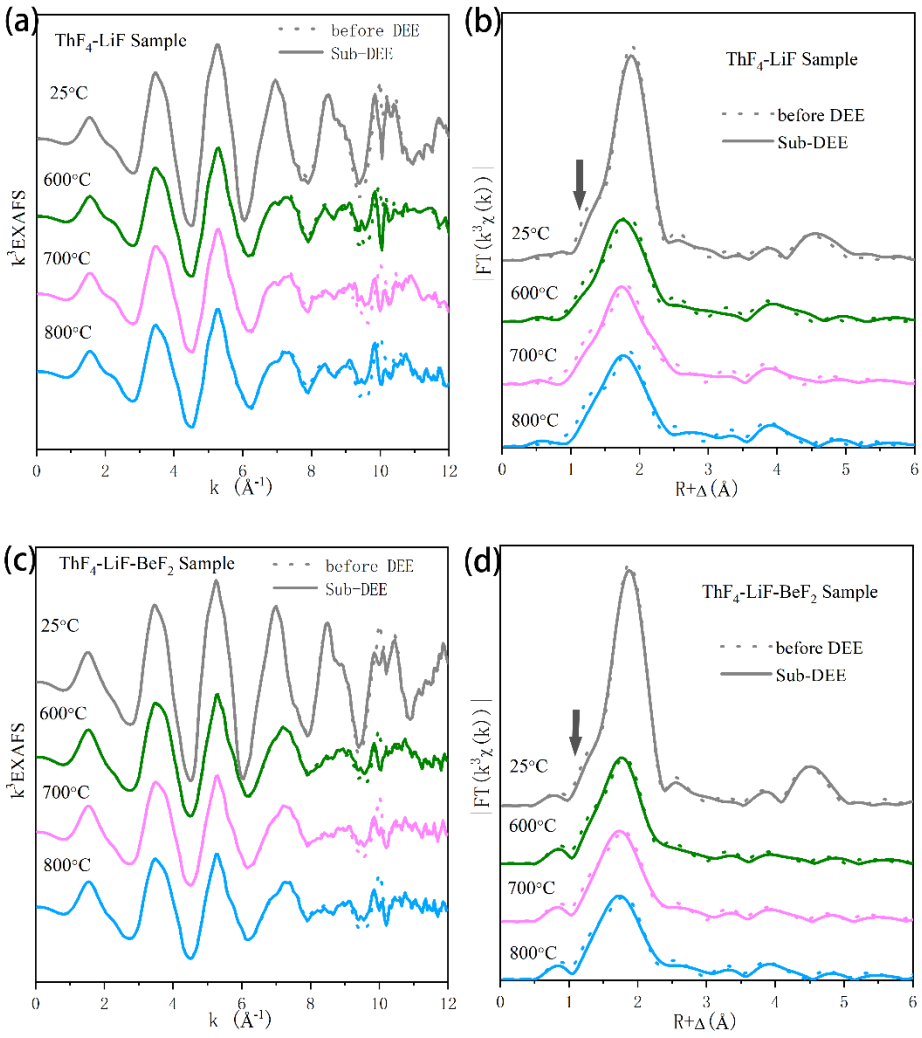

Figure S3 Comparison of experimental thorium L_3-edge k^3-weighted EXAFS oscillation data and Fourier Transform data before and after subtracting the double-electron excitation in ThF_4-LiF and ThF_4-LiF-BeF_2 molten salt vs temperature.

S3. Quantitative local structural information extracted by EXAFS analysis.

Table S1 Structural parameters of Th-F_{1st} bond XAFS at different temperature in the ThF_4-LiF and ThF_4-LiF-BeF_2 compounds from EXAFS fitting.

Sample	Temperature	CN[a]	R(Å)[b]	σ^2(Å^2)[c]	$C_3\times10^{-3}$(Å^3)	R-factor
ThF_4-LiF	25℃	9.0±0.2	2.36	0.009		0.01
	600℃	8.1±0.2	2.33	0.015	2.5±0.2	
	700℃	8.0±0.3	2.33	0.015	3.2±0.4	
	800℃	7.6±0.3	2.33	0.015	3.6±0.4	
ThF_4-LiF-BeF_2	25℃	10.3±0.2	2.35	0.009		0.01
	600℃	8.7±0.2	2.32	0.015	3.4±0.2	
	700℃	8.3±0.3	2.32	0.016	3.7±0.3	

800℃	8.1±0.3	2.32	0.017	3.8±0.4

[a]CN: coordination number. [b]Error: R ≤ ±0.01Å. [c]Debye-Waller factor. Error: $\sigma^2 \leq \pm 0.0005 Å^2$.

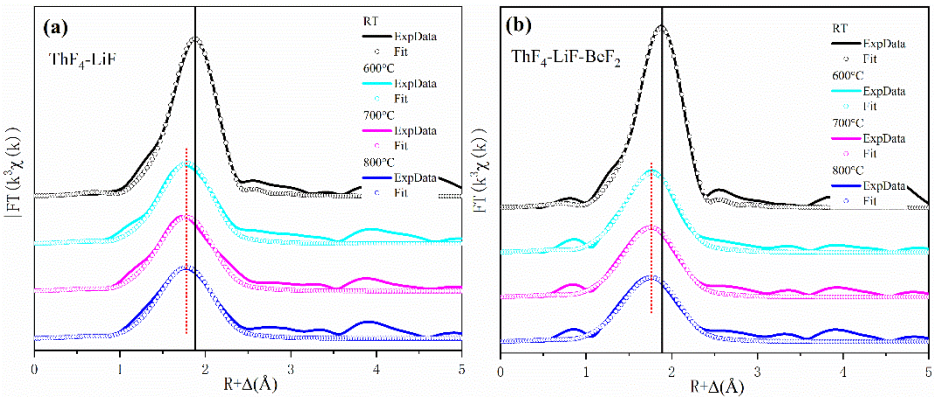

Figure S4 Experimental Fourier transform at the thorium L3-edge EXAFS data of ThF4-LiF and ThF4-LiF-BeF2 compounds and their corresponding fits in R space: —, experimental data; ○, theoretical fit.

S4. Experimental XANES spectra

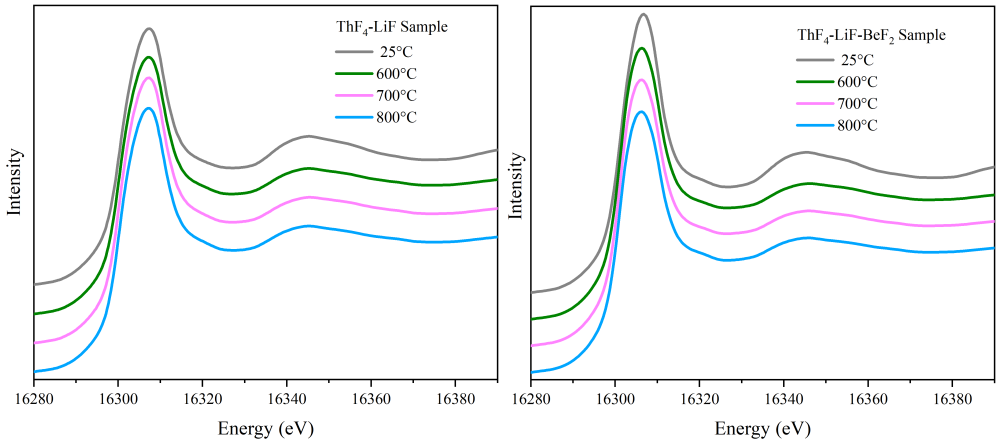

Figure S5 Comparison of experimental thorium L_3-edge XANES spectra in ThF_4-LiF and ThF_4-LiF-BeF_2 molten salt vs temperature.

S5. Cage-correlation-function analysis by MD method.

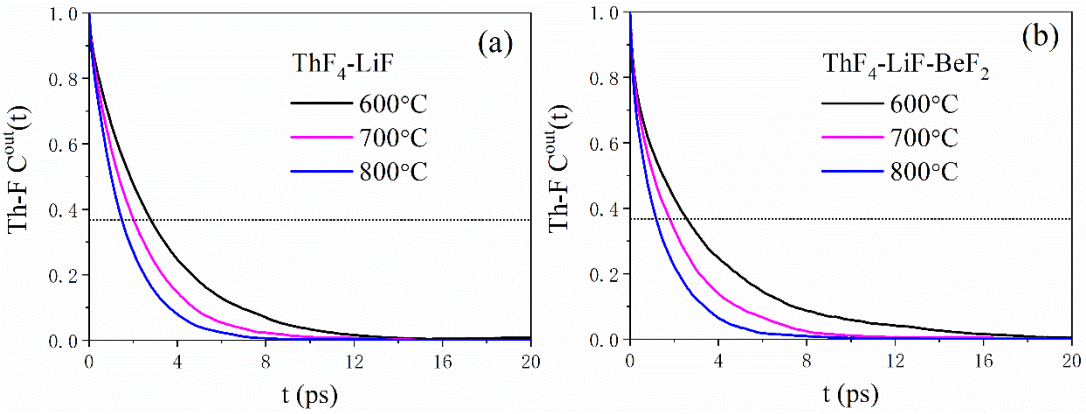

Figure S6 Th-F cage correlation functions in (a) ThF_4-LiF and (b) ThF_4-LiF-BeF_2 systems at different temperatures.