Supplemental Information for Surface Thermodynamics of Yttrium Titanate Pyrochlore Nanomaterials

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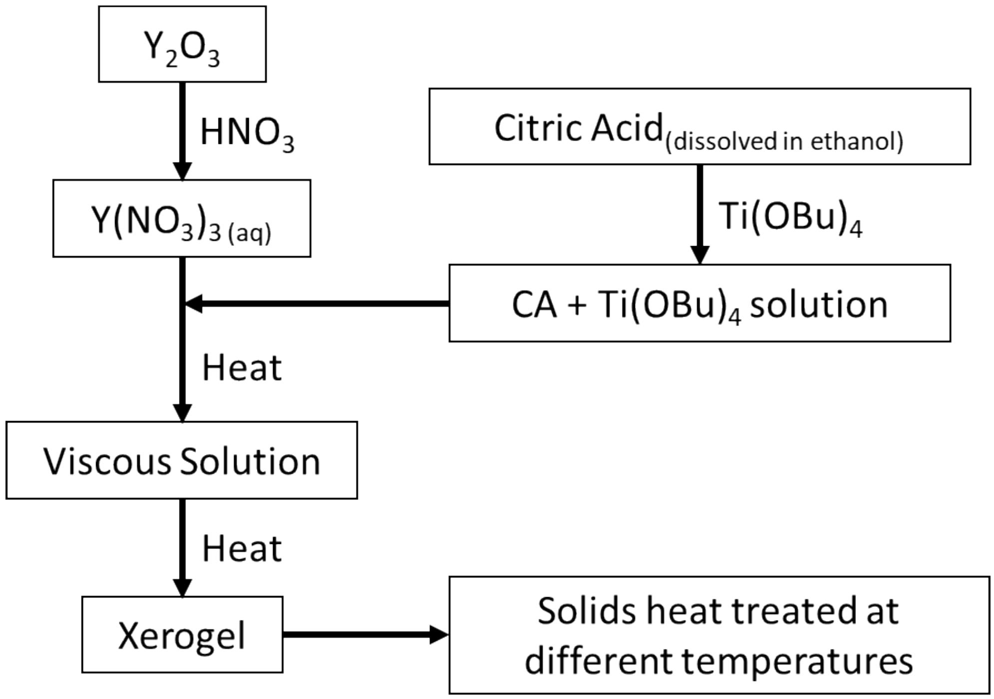


Figure S1. Diagram illustrating the sol-gel synthesis route of as-prepared $Y_2Ti_2O_7$ nanoparticles.

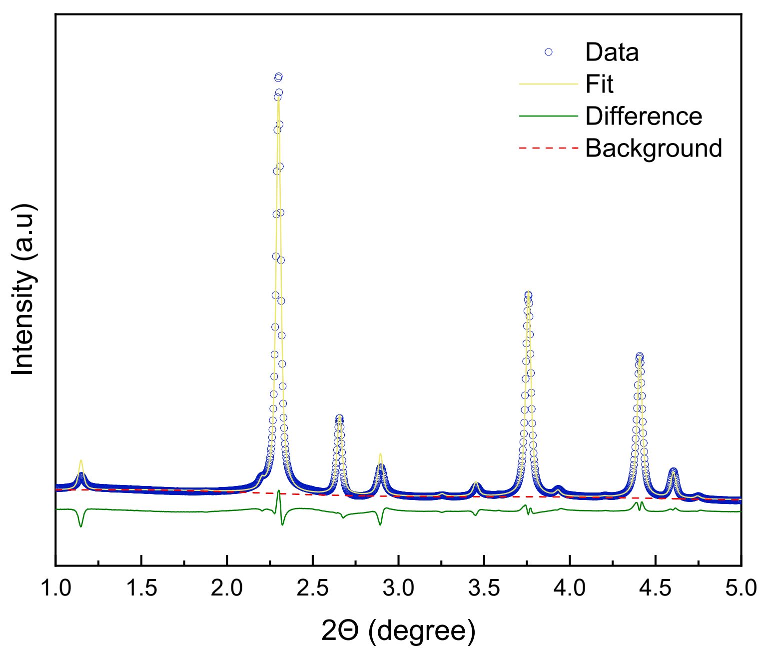


Figure S2. Representative fit of synchrotron XRD pattern of 31 nm sized $Y_2Ti_2O_7$.

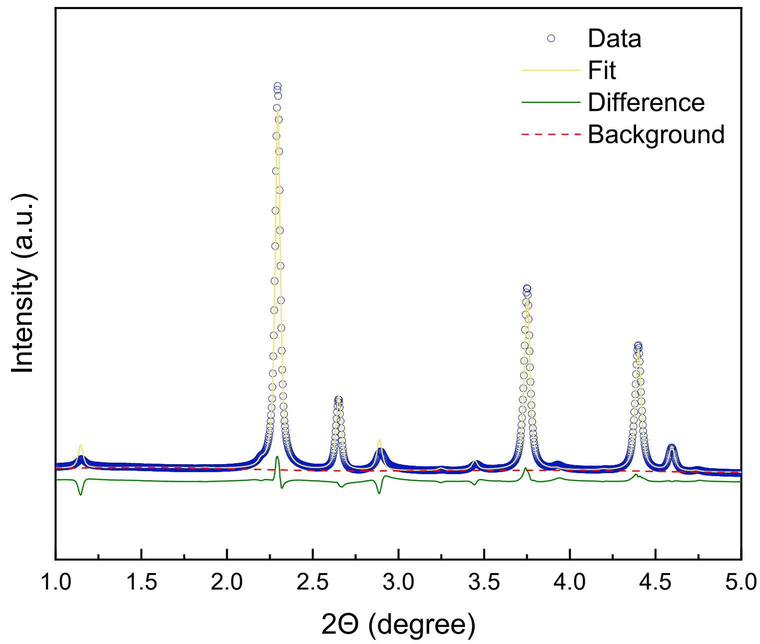


Figure S3. Representative fit of synchrotron XRD pattern of 35 nm sized $Y_2Ti_2O_7$.

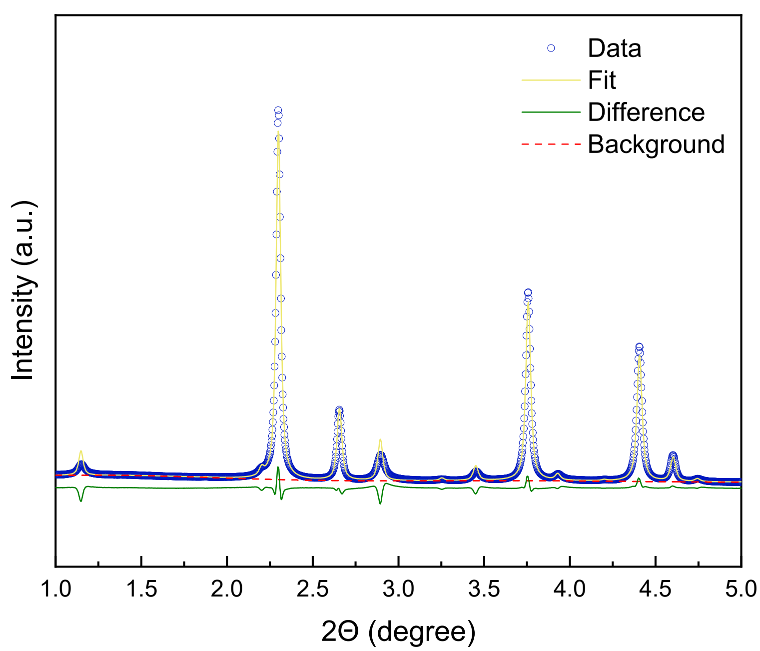


Figure S4. Representative fit of synchrotron XRD pattern of 42 nm sized $Y_2Ti_2O_7$.

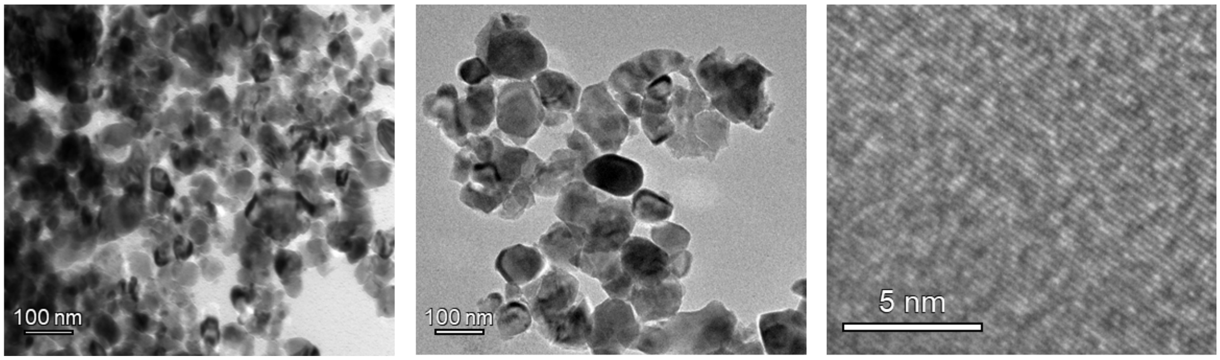
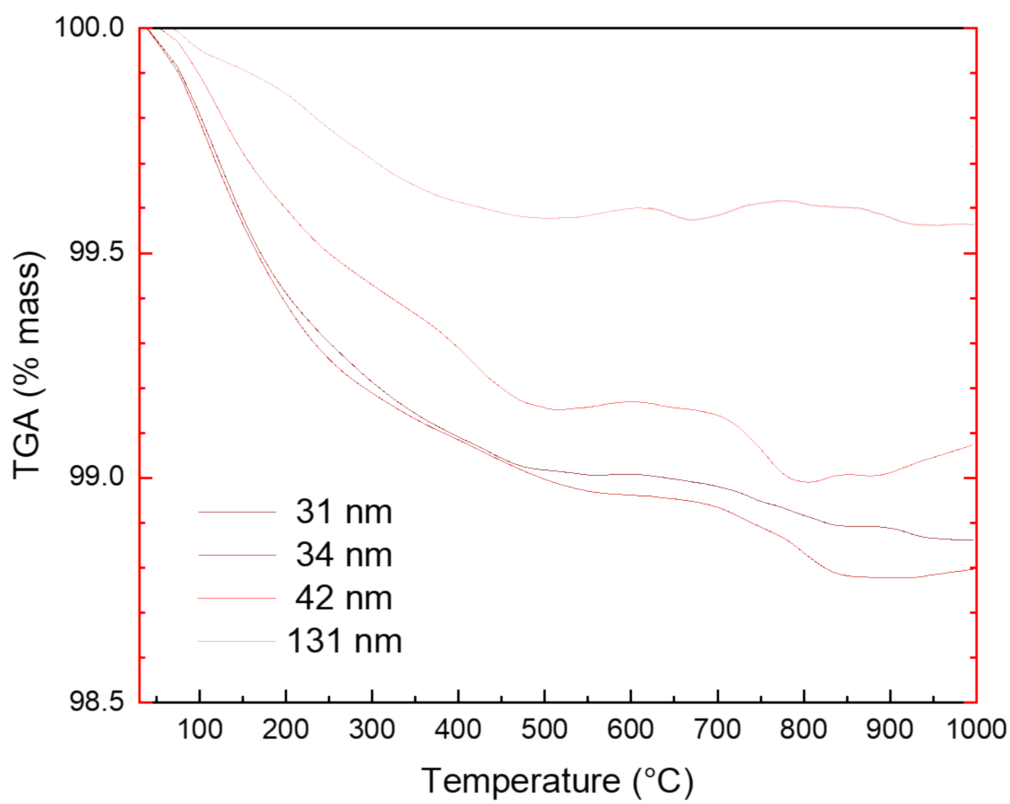


Figure S5. TEM images of the 34 nm (left) and 131 nm (center) nanoparticles and HRTEM of the 131 nm (right) material.



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igure S6. TGA curves under a flowing N_2 atmosphere. Color lightens as the particle diameter increases.

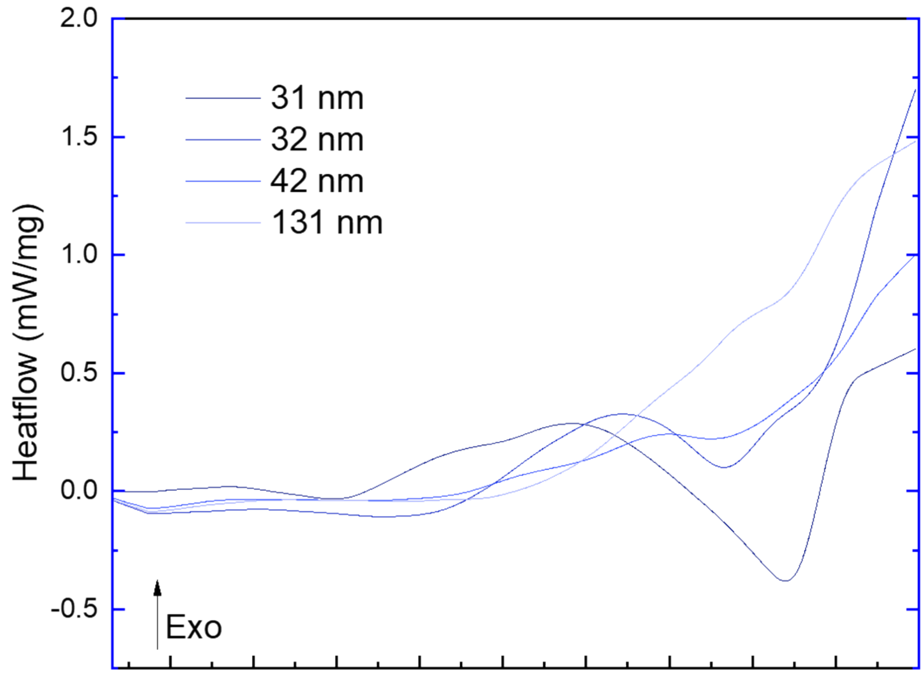

Figure S7. DSC curves under a flowing N_2 atmosphere. Color lightens as the particle diameter increases.

Table S1. Experimentally determined enthalpy of drop solution (ΔH_{ds}), corrected enthalpy of drop solution ($\Delta H_{ds}'$), enthalpy of formation from the oxides ($\Delta H_{f,ox}$) and standard enthalpy of formation (ΔH_f^o)for all pyrochlore samples following the thermochemical cycle in Table 2.

$Y_2Ti_2O_7$ Sample	ΔH_{ds} (kJ/mol)	Corrected $\Delta H_{ds}'$ (kJ/mol)	$\Delta H_{f,ox}$ (kJ/mol)	ΔH_f^o (kJ/mol)
30.83	62.65† ± 0.41‡ (5)§	38.92 ± 0.41	-38.04 ± 0.47	-3858.84 ± 5.46
34.74	56.72 ± 0.57 (3)	32.99 ± 0.57	-32.11 ± 0.61	-3852.91 ± 5.46
41.56	68.17 ± 1.29 (4)	47.83 ± 1.29	-46.95 ± 1.31	-3867.75 ± 5.60
131.4	78.79 ± 1.92 (4)	67.49 ± 1.92	-66.61 ± 1.94	-3887.41 ± 5.78

†Average. ‡Two standard deviations of the average. §Number of measurements.

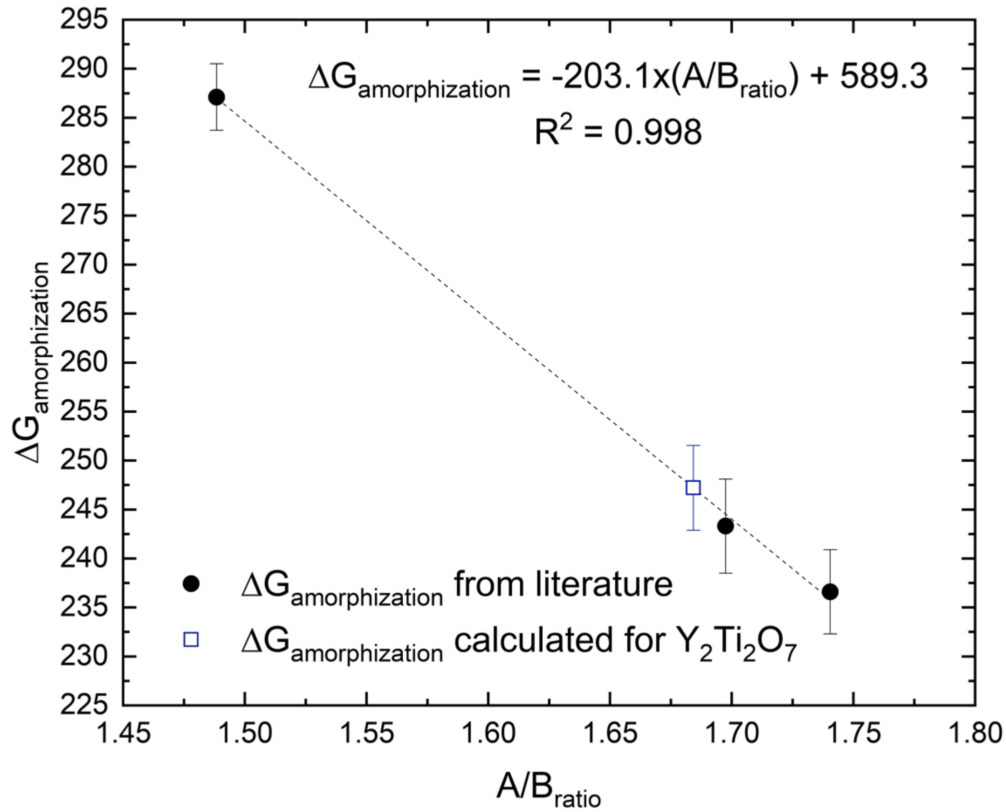

Figure S8. Linear fit of $\Delta G_{amorphization}$ energies of reported pyrochlore compounds[1–3] (black circles) in relation to A/B cationic radii ratio[4]. This fit was used to estimate a value of $\Delta G_{amorphization}$ of $Y_2Ti_2O_7$ (blue square).

References

(1) Chung, C. K.; Lang, M.; Xu, H.; Navrotsky, A. Thermodynamics of Radiation Induced Amorphization and Thermal Annealing of Dy2Sn2O7 Pyrochlore. *Acta Mater* **2018**, *155*, 386–392. https://doi.org/10.1016/j.actamat.2018.06.003.

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(3) Chung, C. K.; O'Quinn, E. C.; Neuefeind, J. C.; Fuentes, A. F.; Xu, H.; Lang, M.; Navrotsky, A. Thermodynamic and Structural Evolution of Mechanically Milled and Swift Heavy Ion Irradiated Er2Ti2O7 Pyrochlore. *Acta Mater* **2019**, *181*. https://doi.org/10.1016/j.actamat.2019.09.022.

(4) Shannon, R. D. Revised Effective Ionic Radii and Systematic Studies of Interatomic Distances in Halides and Chalcogenides. *Acta Crystallographica Section A* **1976**, *32* (5). https://doi.org/10.1107/S0567739476001551.