Supporting information

High-pressure-assisted X-ray-induced damage as a new route for chemical and structural synthesis.

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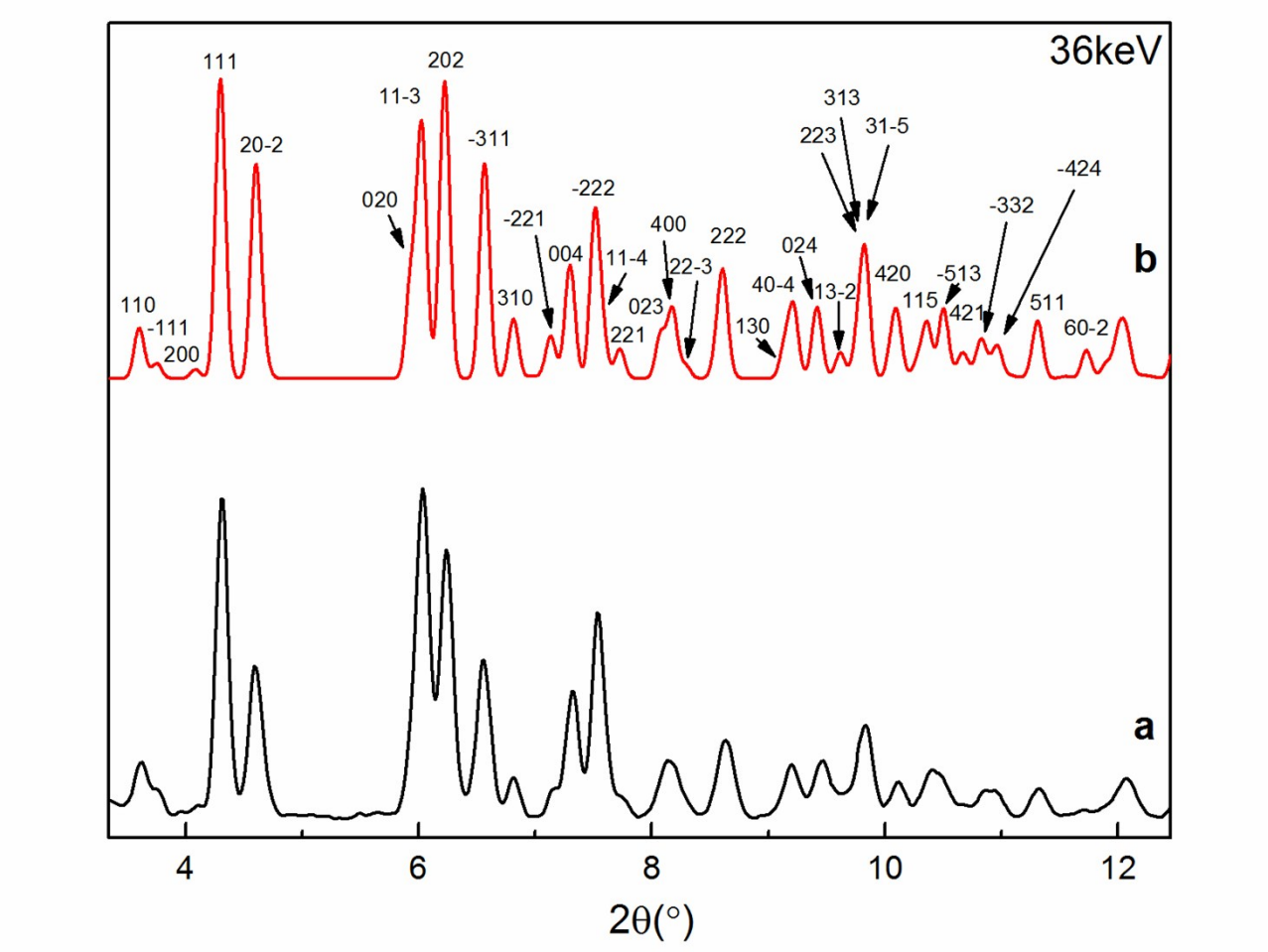

Fig. S1. XRD patterns of cesium oxalate hydrate at ambient conditions at 36 keV X-ray energy. **a**, Experimentally obtained XRD pattern in this work. **b**, XRD pattern of monoclinic crystal structure of cesium oxalate hydrate[1] with a space group *C2/c* and lattice parameters: a = 10.1127 Å, b = 6.6535 Å, and c = 11.3257 Å. Diffraction peaks are labeled with corresponding Miller indices.

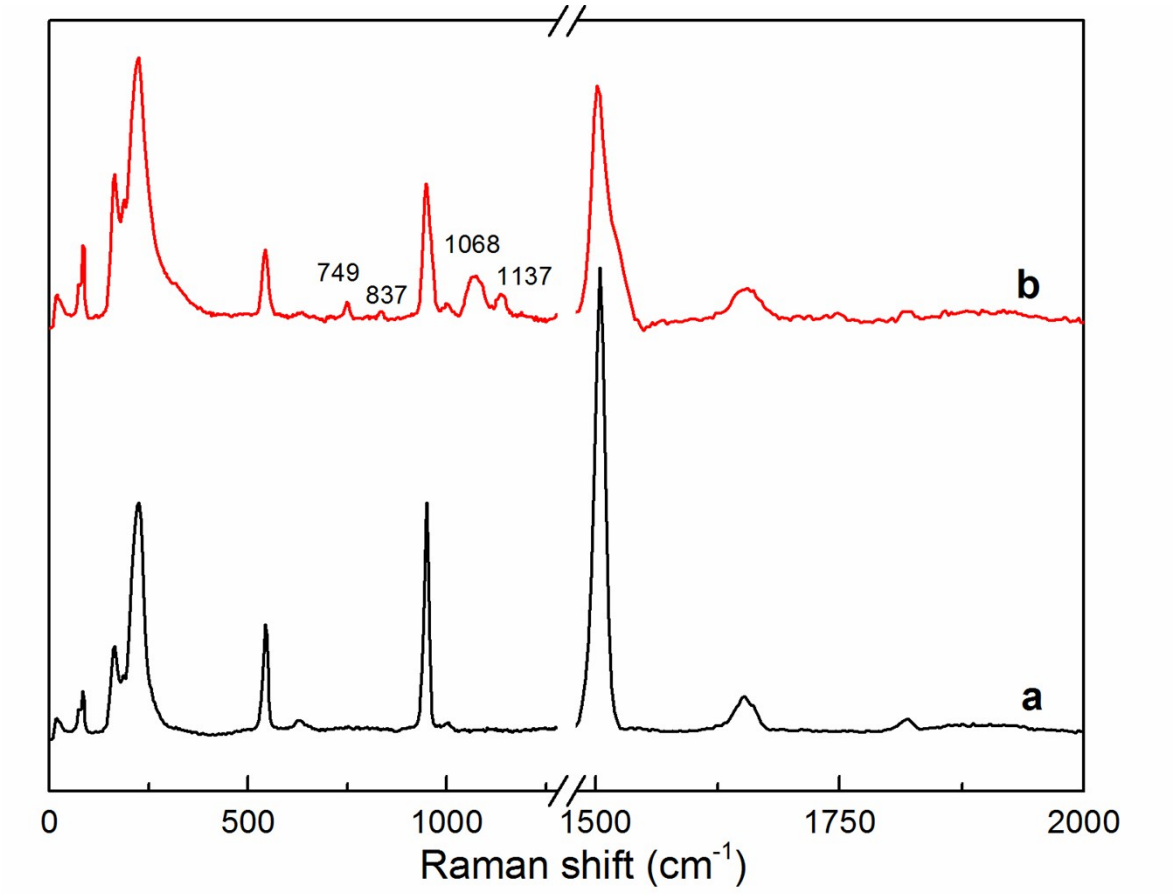

Figure S2. Raman spectra of cesium oxalate hydrate used in our experiments. a, Unirradiated/virgin point in the sample. **b**, Damaged/irradiated spot on the same sample. Assignments of selected peaks are presented in Table S1.

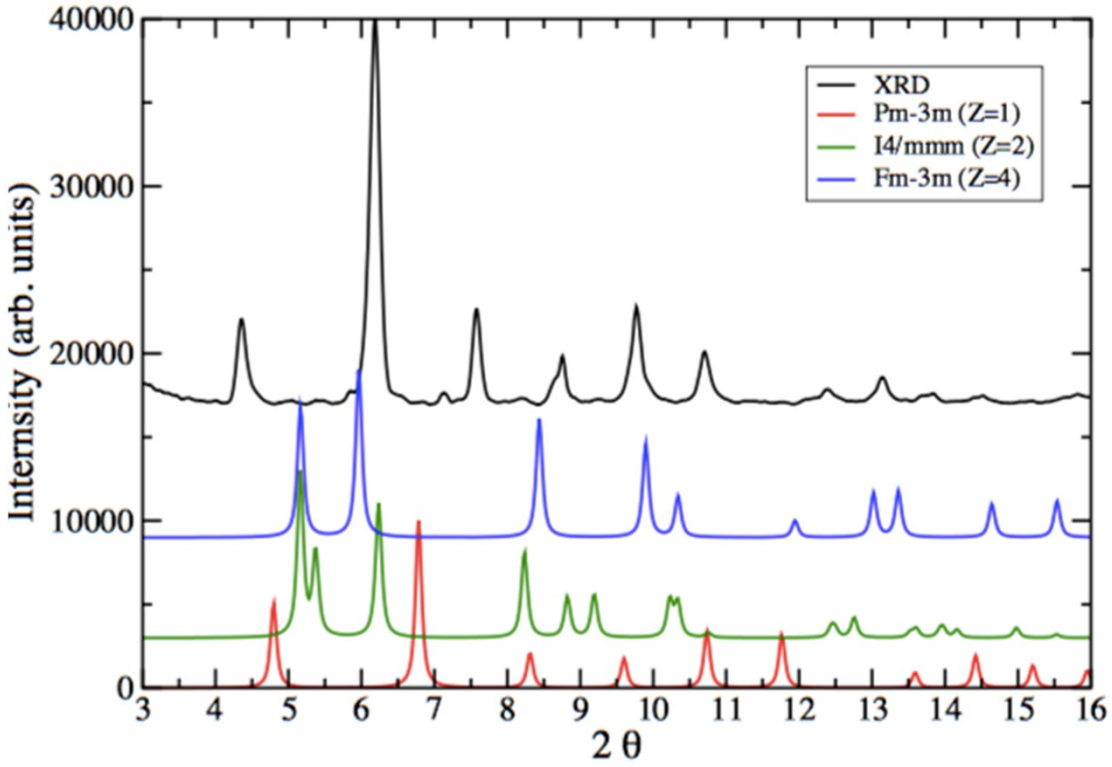

Fig. S3. Simulated XRD patterns of the optimized CsO_2 structures in Pm-3m (Z=1), I4/mmm (Z=2), and Fm-3m (Z=4). Total and cohesive energies of three CsO_2 polymorphs are reported in Table S2.

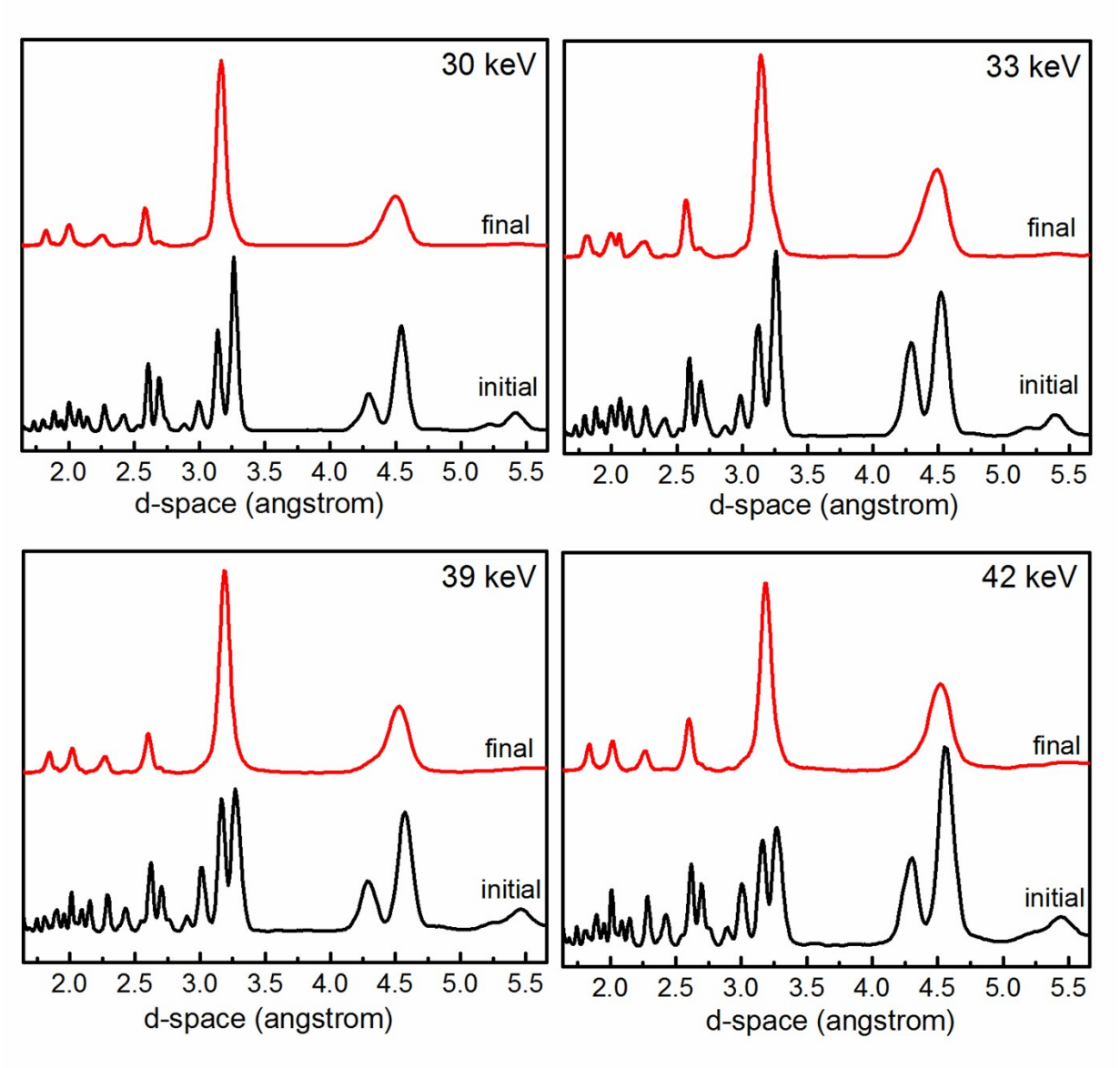

Fig. S4. XRD patterns of cesium oxalate hydrate before (initial) and after 40 min (final) of X-ray irradiation at different energies.

Table S1. Selected vibrational bands and assignments of cesium oxalate hydrate after 40 min of X-ray irradiation at 0.3 GPa.

Peaks position, cm^{-1}	Vibrational mode assignment (ref)
749	Vibration of O_2^{2-} in Cs_2O_2 (ref. 2,3)
837	Cesium hydrate impurities (ref. 2,3)
1068	COO rocking(C-H bending out-of-plane) in $CHCsO_2$ (ref. 4)
1137	Vibration of O_2^- in CsO_2 (ref. 2,3,5-7)

Table S2. Calculated total and cohesive energies of three CsO_2 polymorphs.

Space group	Total energy (eV/atom)	Cohesive energy (eV/atom)
Pm-3m (Z=1)	-4.340	2.835
I4/mmm (Z=2)	-4.396	3.002
Fm-3m (Z=4)	-4.339	2.833

References

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