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The Effect of Ionizing Radiation on U^{6+} Phases

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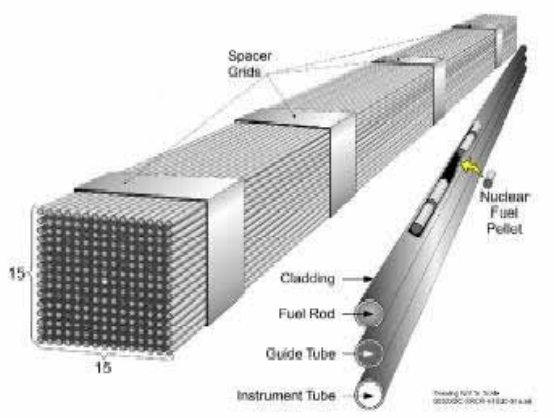
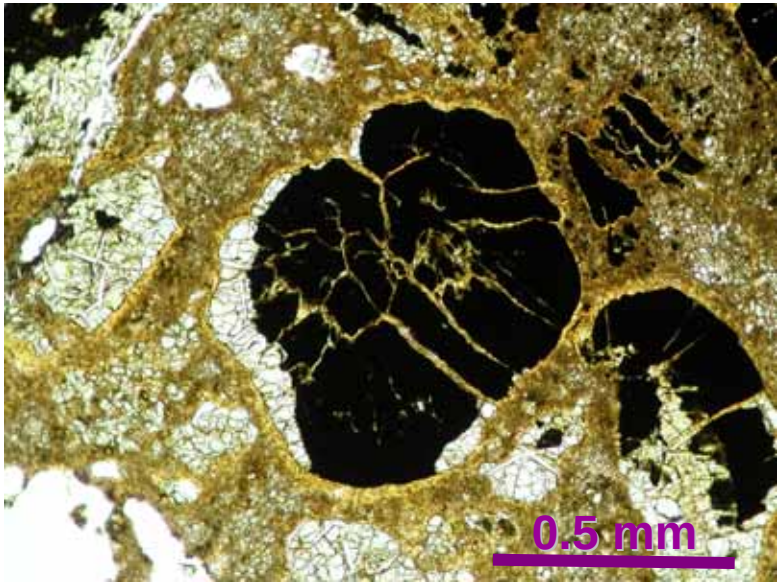
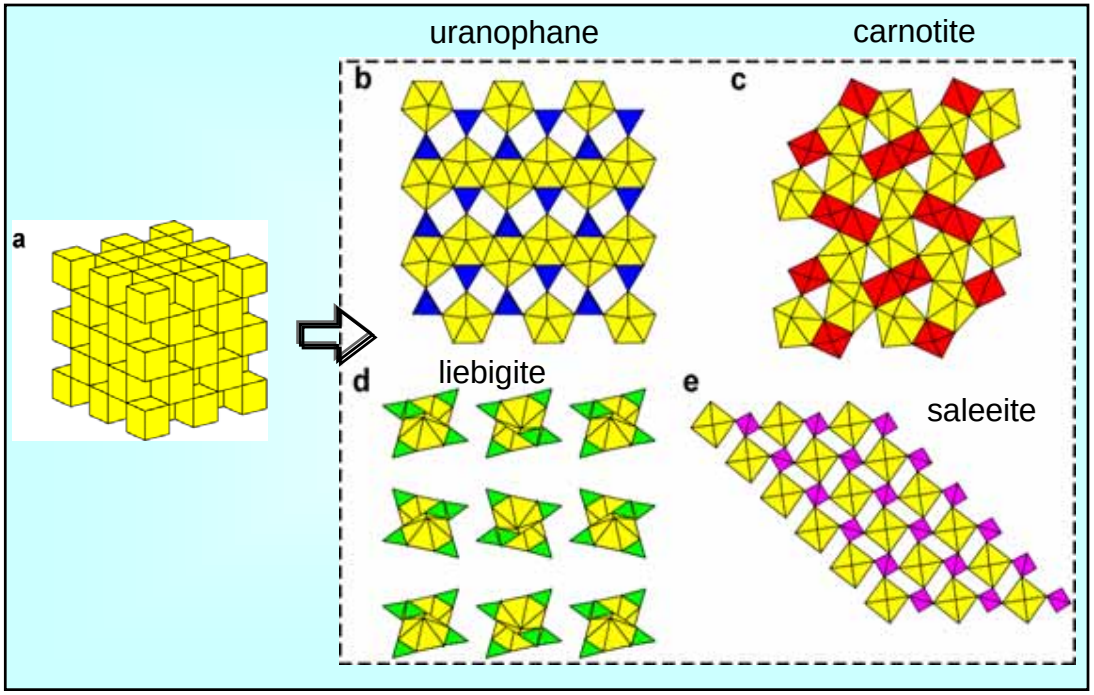
Introduction

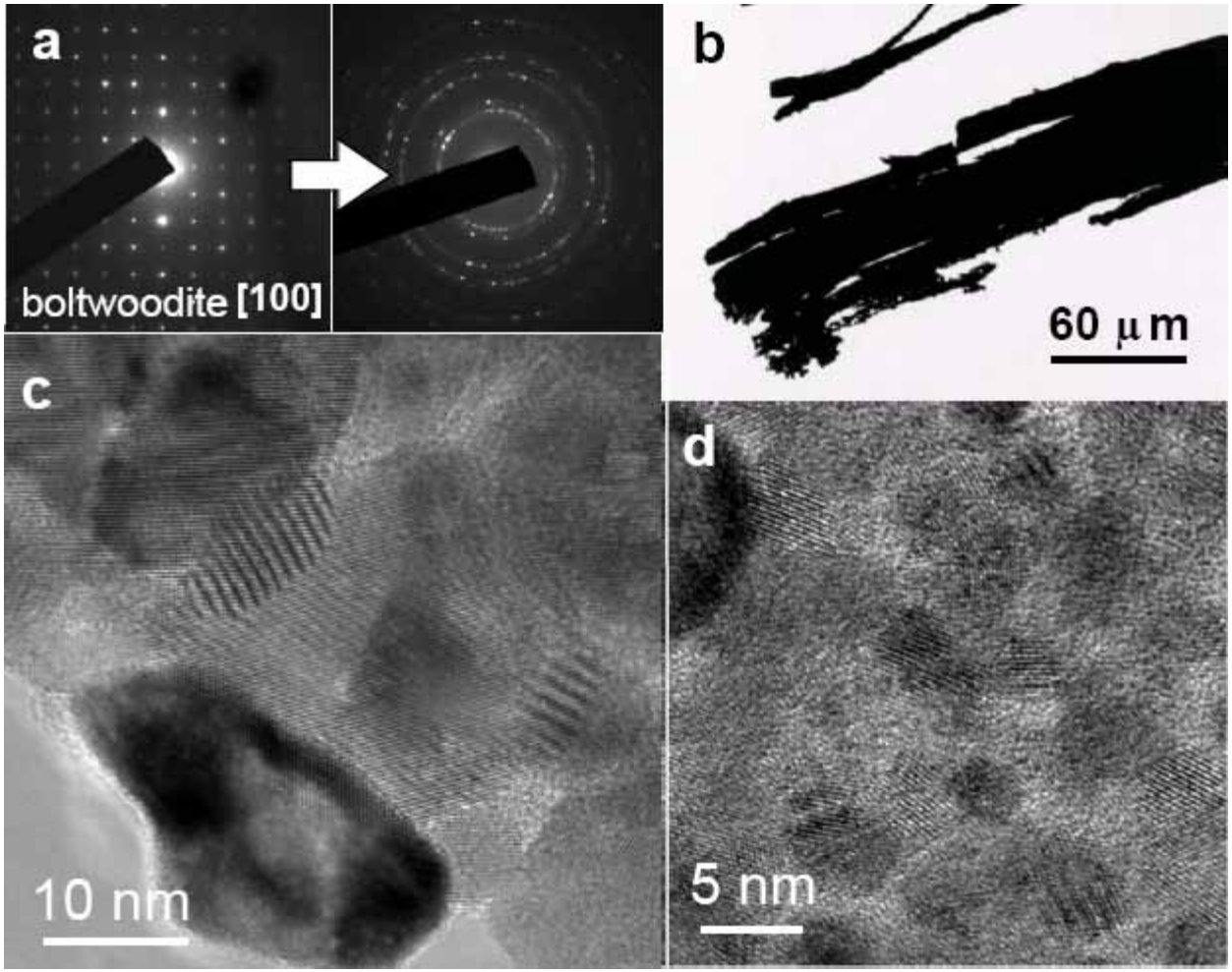






Effect of ballistic interaction in U^{6+} phases

boltwoodite, 1.0 MeV Kr^{2+}, room temperature



Electron irradiation of U^{6+}-minerals

Goal

To evaluate the stability of U^{6+}-phases under ionizing radiation.

Materials

boltwoodite	$K[(UO_2)(SiO_3OH)](H_2O)_{1.5}$
kasolite	$Pb[(UO_2)(SiO_4)](H_2O)$
saléeite	$Mg[(UO_2)(PO_4)]_2(H_2O)_{10}$
carnotite	$K_2(UO_2)_2(V_2O_8)(H_2O)_3$
liebigite	$Ca_2[(UO_2)(CO_3)_3](H_2O)_{11}$
schoepite	$[(UO_2)_8O_2(OH)_{12}](H_2O)_{12}$
uranophane	$Ca[(UO_2)(SiO_3OH)]_2(H_2O)_5$
sklodowskite	$Mg[(UO_2)(SiO_3OH)]_2(H_2O)_6$

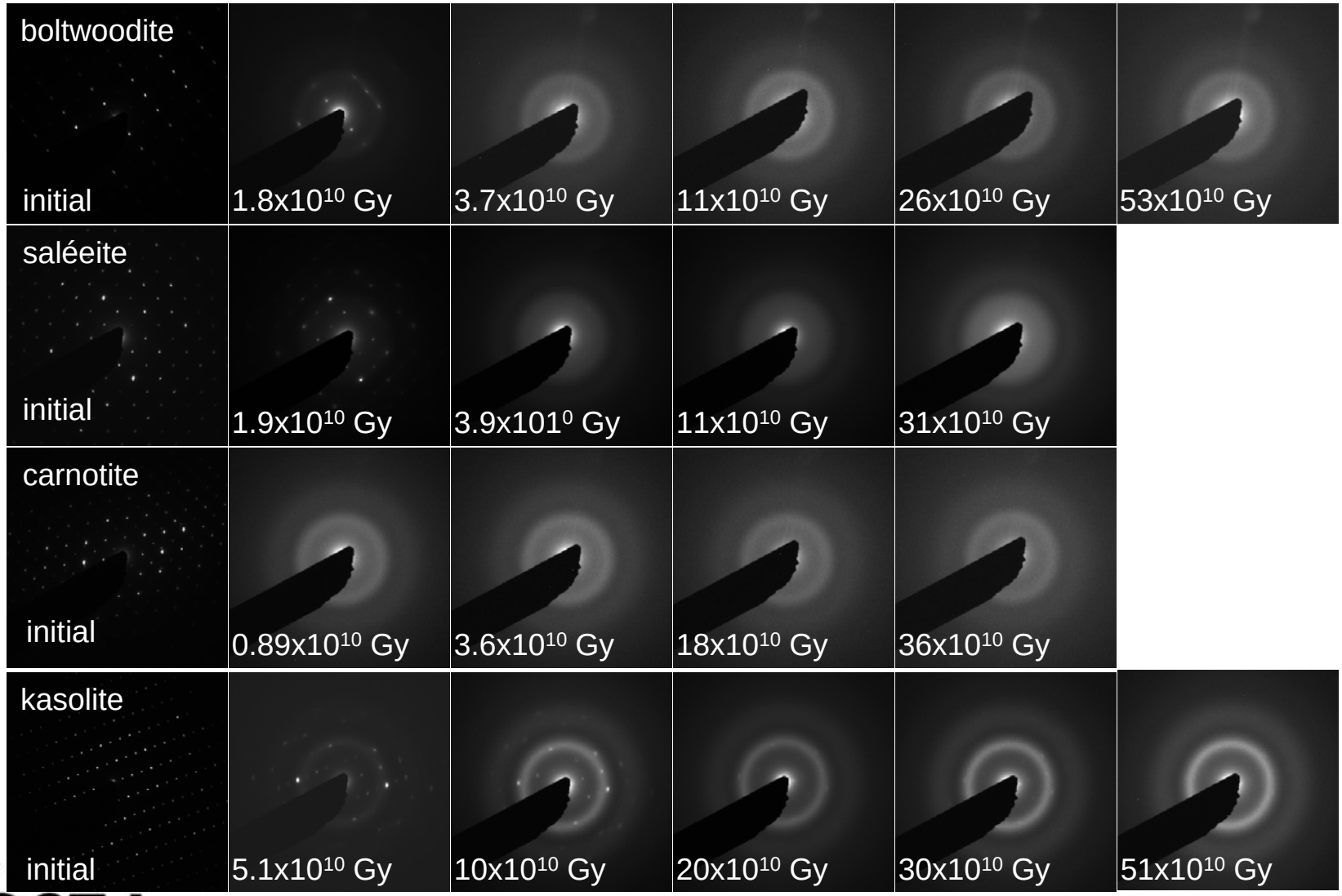
→ Utsunomiya et al. (2003) Am. Mineral.

Experimental Methods

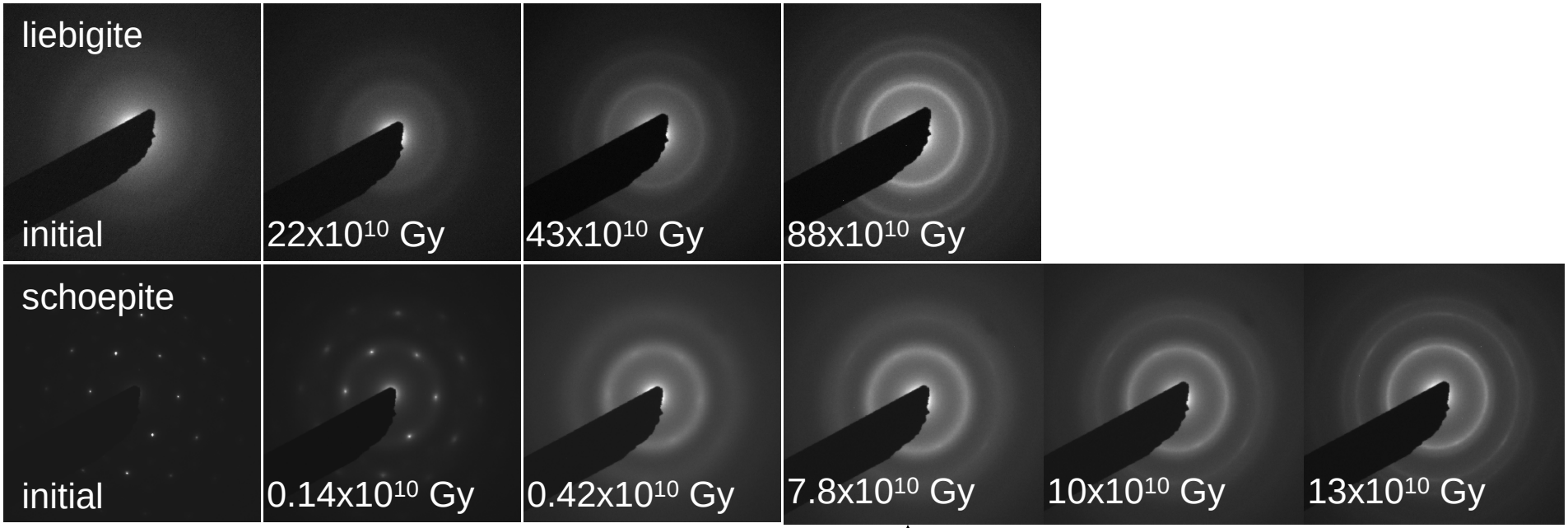
- **Electron irradiation (ionizing radiation) was performed using JEOL 2010F at EMAL of University of Michigan.**
  - **Structural transition was monitored in situ using selected area electron diffraction pattern (SAED).**
  - **The transition in HRTEM was also recorded on video.**
  - **Temperature: room temperature and 300 ^{o}C**
  - **e-flux = ~8-33 x 10^{17} (e^{-}/cm^{2}/sec),**
- **SRIM 2003 (The Stopping and Range of Ions in Matter) calculation (Ziegler et al., 2003)**

SAED of transition during e-irradiation

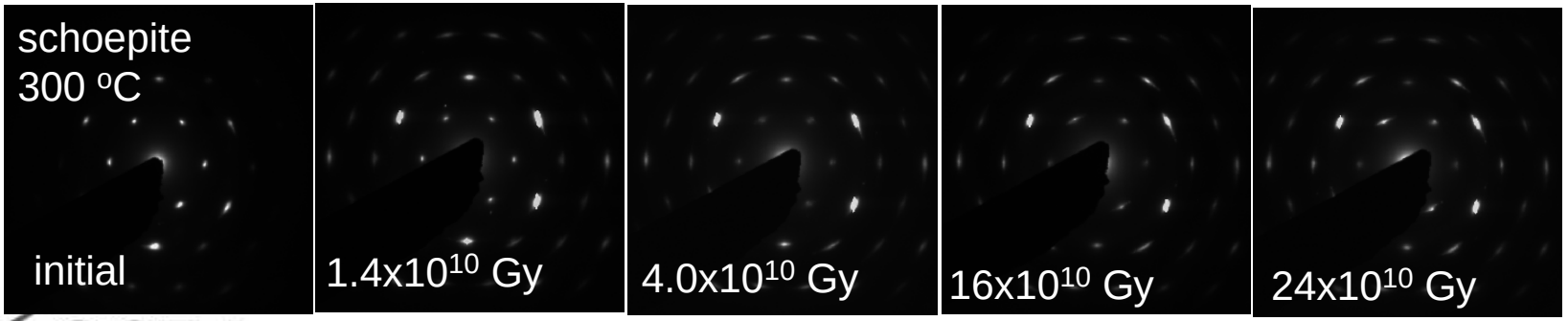
e-flux = ~8-33 x 10^{17} (e-/cm^2/sec), room temperature



SAED of transition during e-irradiation



uraninite nanocrystals start to form

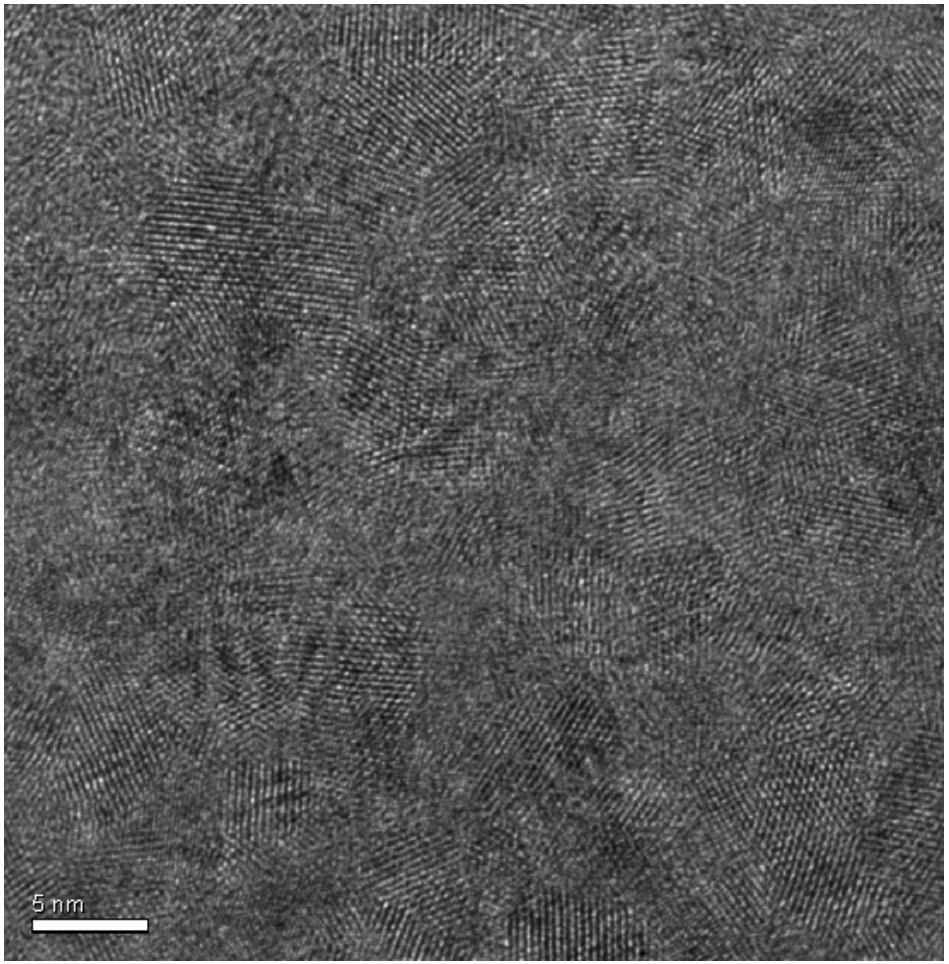


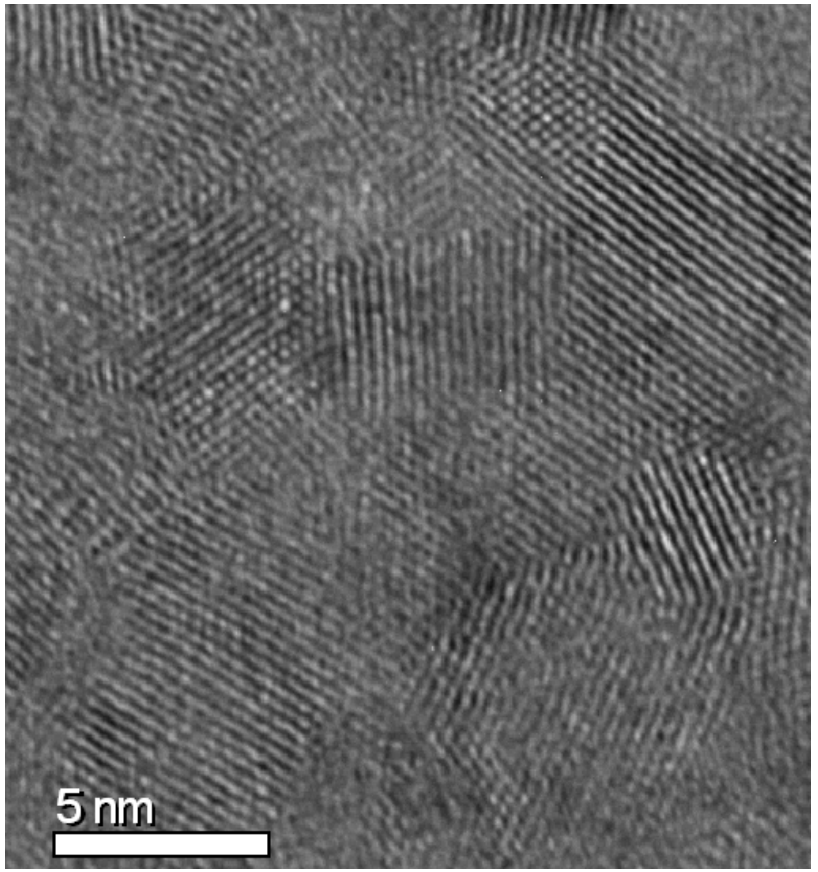
HRTEM of transition on video

In situ observation for nanocrystallization in schoepite during electron irradiation at room temperature

Video: schoepite hrtem10

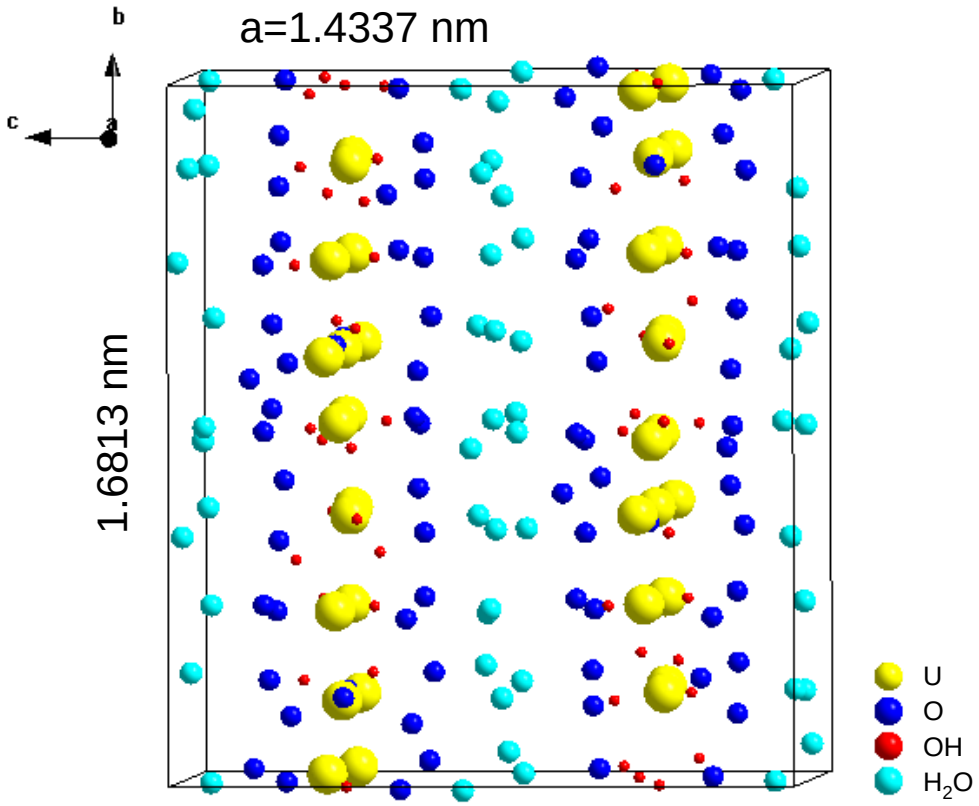
HRTEM image of nanocrystalline uraninite formed after ~13x10^{10} Gy of the electron fluence.





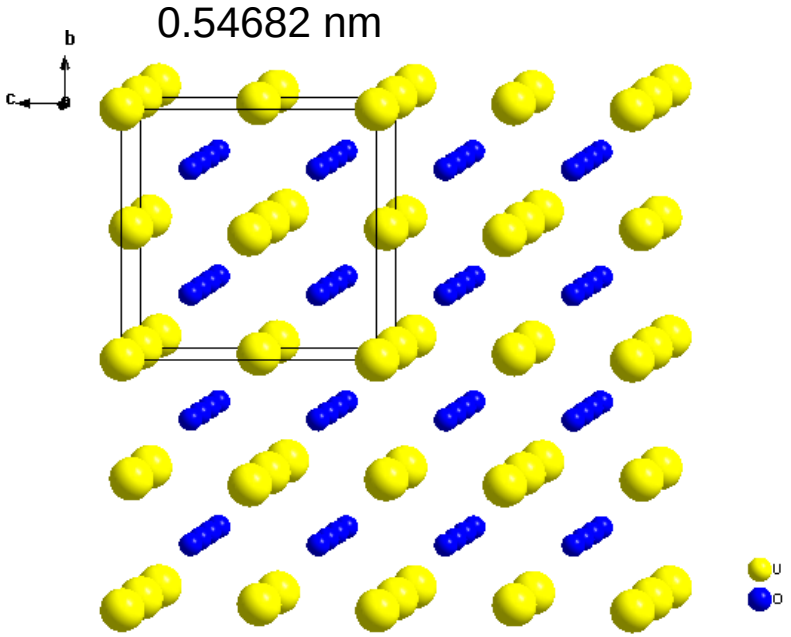
Comparison of the structures

schoepite



U-O: 0.1780 nm & 0.2280 nm
U-OH: 0.2420 nm
U-U: ~0.4 nm (0.3830-0.4570)

uraninite



U-O: 0.2370 nm
U-U: 0.3870 nm

Conclusions (1)

- During the irradiation, boltwoodite, saléeite, and carnotite became amorphous at doses of 1-4 x 10^{10} gray (Gy), while the amorphization dose (D_c) of kasolite, 50x10^{10} Gy, was about an order of magnitude higher than that of boltwoodite. This high D_c for kasolite is consistent with the hypothesis that the D_c increases as the mass of the inter-layer cation increases.
- Only amorphization, rather than chemical decomposition, occurred in boltwoodite, saléeite and carnotite, even at doses as high as 80x10^{10} Gy.
- In contrast, uraninite nanocrystallites began to form with a random orientation at ~43x10^{10} Gy in liebigite that had already become amorphous prior to irradiation in the vacuum of the TEM.

Conclusions (2)

- For schoepite, the D_c was only 0.51×10^{10} Gy, and randomly oriented uraninite nanocrystallites began to form at 7.8×10^{10} Gy, which is approximately the same as compared with the D_c of the other U^{6+}-phases.
- All of these doses are higher than the predicted cumulative dose by the ionizing radiation in spent nuclear fuel (~10^7-10^8 Gy during the first 10^{2-3} years after discharge).
- The effect of electron irradiation during the ion irradiation experiments was negligible.