Determination of Transmutation Effects in Crystalline Waste Forms

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Abstract

Small sealed stainless steel capsules containing pollucite ($CsAlSi_2O_6$) have been in storage for over 20 years. During this time, the ^{137}Cs has been decaying to ^{137}Ba, which is not radioactive. This decay places the pollucite structure under significant strain because of the limited solubility of Ba in pollucite. We have been examining this material with the hope of characterizing the effects of the transmutation from Cs to Ba on the pollucite structure and the fate of the Ba in the solid. Originally, we hoped to use x-ray absorption fine structure (XAFS) to characterize the material without opening the capsules. Initial data suggest that this approach is not possible, although a couple of additional approaches will be taken at the Argonne Advanced Photon Source Facility. Instead, we have elected to open the capsules carefully in an inert gas glove box. Portions of the sample from the capsules will be used for XAFS, nuclear magnetic resonance (NMR), and transmission electron microscopy (TEM). Of these techniques, both TEM and NMR appear to be yielding the best results. We will still attempt to use EXAFS to determine if the Ba exists as Ba metal or Ba oxide inside the capsules.

Pollucite capsule, with dime, for scale. This capsule contained only nonradioactive cesium, despite the stamp.

Background

Originally fabricated as radiation sources for tumor treatments, the samples are sealed, 300 micrometre-thick, stainless steel capsules that are approximately 3 mm long and 1 mm in diameter, and contain approximately 8.0 mg of pollucite. These samples of pollucite range in age from 7 years to 24 years old. Additional samples are available in which no ^{137}Cs was incorporated. Baseline data are being collected on these nonradioactive samples. Inside the capsules is a loosely consolidated pellet of pollucite. This material appears to be a pure phase when examined with XRD, but some secondary phase is found with the TEM.

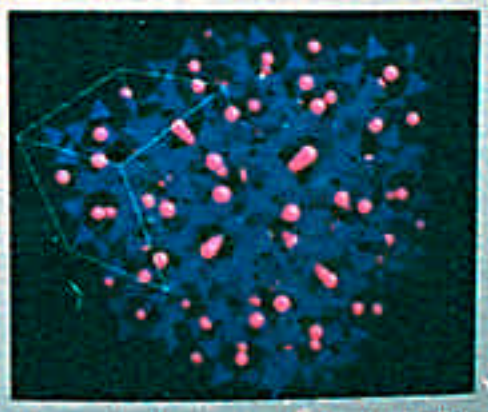
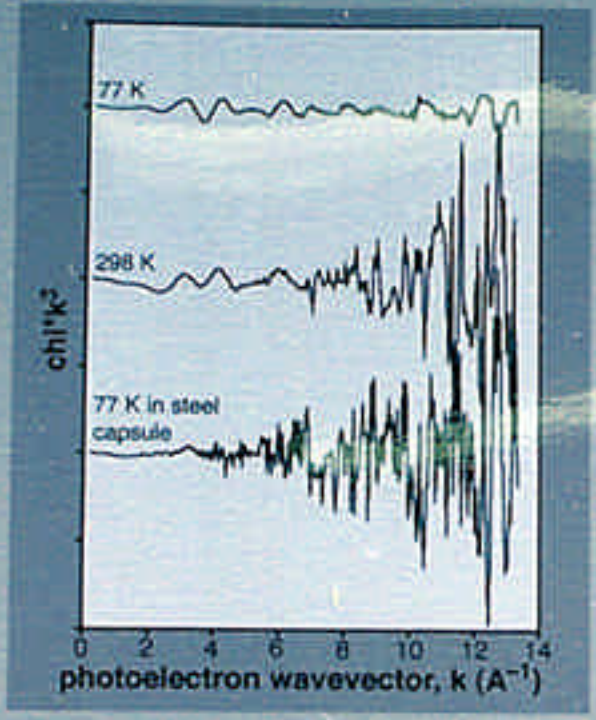
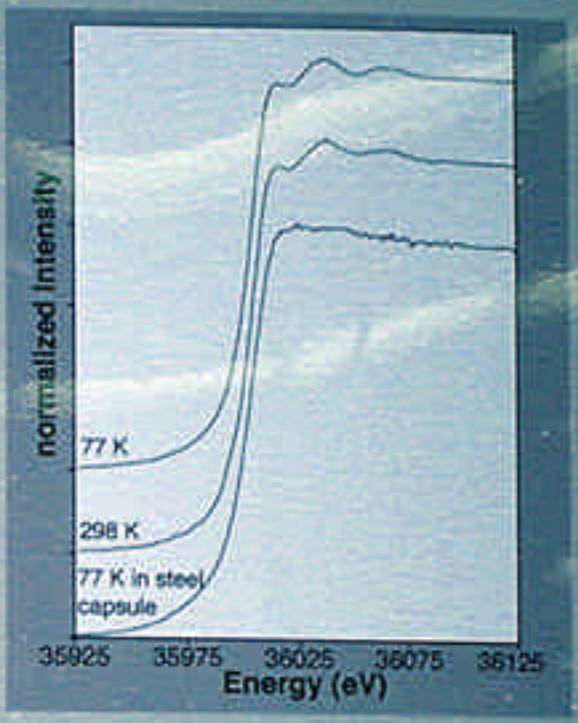


Image of pollucite crystal structure. The tetrahedra are either AlO_4 (1/3) or SiO_4 (2/3).

Results

EXAFS

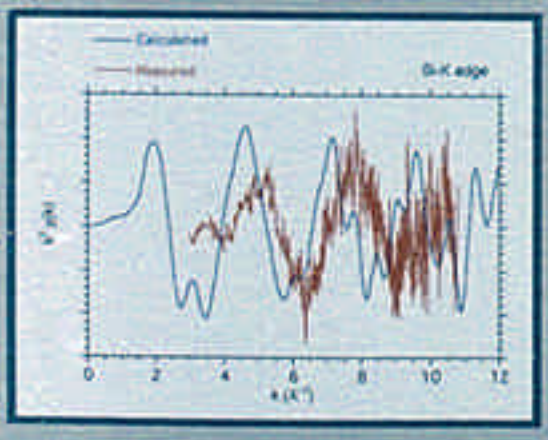




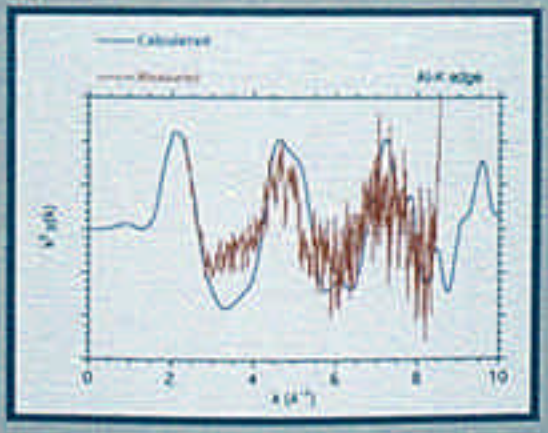
Results show that the EXAFS of Cs in these complex minerals is weak and difficult to interpret when obtained through stainless steel at 77 K.

TEM

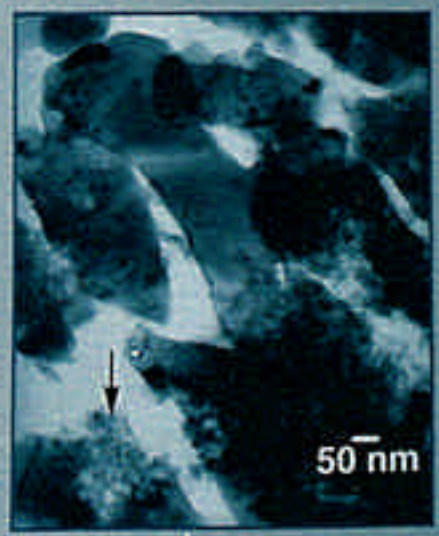
(a)



(b)



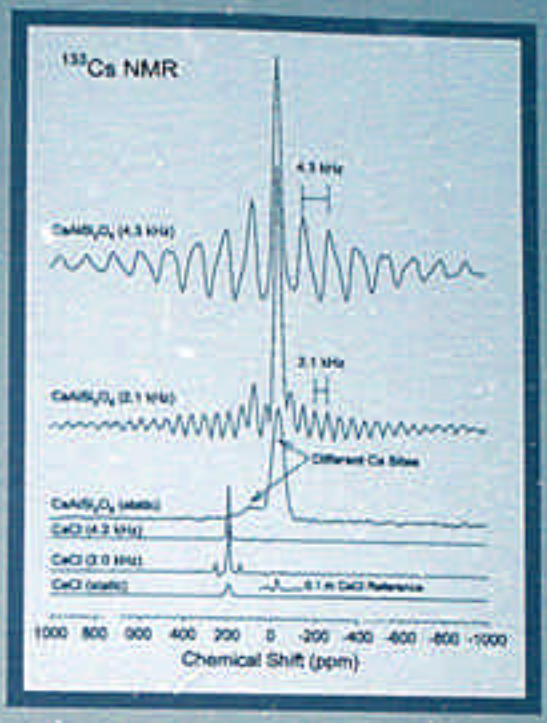
(a) Comparison of calculated (left) and experimental (right) EXELFS from the Si-K edge of pollucite. The Fourier transforms of the experimental $k^2\chi(k)$ appear in (b) for the pollucite. No corrections for phase shift or edge position were made in the EXELFS extracted from the experimental data.



TEM image of pollucite from the nonradioactive capsule

NMR

NMR from the pollucite sample (right) along with reference materials. The spinning frequency for magic angle NMR is indicated in parentheses.



Conclusions

- If Ba exists as metal or oxide, we should be able to determine the state of Ba in the capsules.
- A second phase has been located in the pollucite powder. Parameters for examining the radioactive samples have been determined.
- The second phase has been confirmed with NMR.
- NMR appears to be a promising technique for determining the chemical state of Cs and Ba in these powders.

Future Work

- EXAFS will be used to determine if Ba exists in the capsules as Ba metal or BaO
- Powder will be removed from the capsules and examined with EXAFS, TEM, and NMR.

Acknowledgements

Funded by DOE through the Environmental Management Science Program, under contract W-31-109-ENG-38