

Annual Report for FY 1997

Determination of Transmutation Effects in Crystalline Waste Forms

D. M. Strachan, E. C. Buck, and J. A. Fortner
Argonne National Laboratory

N. J. Hess
Pacific Northwest National Laboratory

Executive Summary

A team from two national laboratories is studying transmutation effects in crystalline waste forms. Analyses are being done with 18 year old samples of ¹³⁷Cs-bearing pollucite ($\text{CsAlSi}_2\text{O}_6 \cdot 0.5\text{H}_2\text{O}$) obtained from a French company. These samples are unique in that the pollucite was made with various amounts of ¹³⁷Cs, which was then sealed in welded stainless-steel capsules to be used as tumor irradiation sources. Over the past 18 years, the ¹³⁷Cs has been decaying to stable Ba in the capsules, i.e., in the absence of atmospheric effects. This material serves as an analogue to a crystalline waste form in which such a transmutation occurs to possibly disrupt the integrity of the original waste form.

Work this year consisted of determining the construction of the capsule and state of the pollucite in the absence of details about these components from the French company. We have opened one capsule containing nonradioactive pollucite. The information on the construction of the stainless-steel capsule is useful for the work that we are preparing to do on capsules containing radioactive pollucite. Microscopic characterization of the nonradioactive pollucite revealed that there are at least two compounds in addition to pollucite: a Cs-silicate and a Cs-aluminosilicate (CsAlSiO_4). These findings may complicate the interpretation of the planned experiments using X-ray absorption spectroscopy. Electron energy loss spectroscopy and energy dispersive X-ray spectroscopy (fluorescence) have been used to characterize the nonradioactive pollucite.

We have investigated the stability of the nonradioactive pollucite to β radiation damage by use of 200 keV electrons in a transmission electron microscope. The samples were found to become amorphous in less than 10 minutes with loss of Cs. This is equivalent to many more years of β radiation damage than under normal decay of the ¹³⁷Cs. In fact, the dose was equivalent to several thousand years of normal radiation damage from the decay of ¹³⁷Cs. Of course, there would not be any ¹³⁷Cs remaining after that length of time because the half-life of ¹³⁷Cs is 30 y.

Preparations have been started to study the radioactive pollucite samples at the Stanford Synchrotron Radiation Laboratory. Our calculations show that by thinning the base of the capsules we should be able to obtain about a factor of ten increase in the fluorescence signal. Procedures for thinning capsules containing the radioactive pollucite and examining the samples at the Stanford synchrotron are in place.

Introduction

At the Hanford site, the high-level nuclear waste that is stored in large underground tanks will be retrieved and processed. A high-level waste stream containing the bulk of the insoluble chemicals from the chemical processing and the bulk of the radionuclides including the

transuranics will result from the processing of these wastes. A low-level waste stream that will constitute the majority of the soluble chemicals that are in the waste and sufficiently small amount of the radionuclides to allow the waste to be classified as less than Class C low-level waste. As part of the processing of the waste in the tanks, ^{137}Cs , ^{90}Sr , and possibly ^{99}Tc will be separated from the wastes. Although the current chemical processing scheme calls for combining these separated radionuclides with the processed high-level waste stream at the vitrification facility, there may be a need to immobilize these radionuclides as separate waste forms because of the problems they cause in vitrification. For ^{137}Cs , relatively high volatility during the vitrification process makes it undesirable to add processing stream high in ^{137}Cs to the vitrification process. Therefore, it may be more cost effective to process the ^{137}Cs -rich effluent from the waste treatment facility to a unique waste form.

Potential waste forms for ^{137}Cs storage include crystalline silicotitanate (KLAVATTER 1994), pollucite ($\text{CsAlSi}_2\text{O}_6$) silicate glass, glass-ceramics, and cesium-loaded zeolites (STRACHAN and SCHULZ 1979). Of these candidates, pollucite offers the advantage of 40 mass% Cs content in a relatively dense mineral. The measured and calculated solubility of pollucite are approximately three orders of magnitude less than that measured for the silicate glass (STRACHAN and SCHULZ 1979 and ANCHELL et al. 1993). Pollucite can be produced from aqueous solutions with readily available materials (STRACHAN and SCHULZ 1979) or by low temperature methods (HOGAN and RISUD 1991).

A concern common to candidate Cs waste forms is the effect of beta radiation and transmutation on the mobility of Cs and on the integrity and safety of the waste form. Most radiation effects work of this type has focused on alpha decay and the associated recoil nuclei of the actinide elements which produce several orders of magnitude more displacements than beta radiation. However, an aspect of the beta decay associated with the fission products is transmutation that results in both a relatively large change in valence and ionic radius, both of which may not be readily accommodated in the host mineral. In pollucite, for example, the structure can accommodate more than 40 wt. % Cs, but less than 10 mass% Ba (LANGLET 1969). The long-term effects of ^{137}Cs transmutation have not been studied previously in any waste form because of its 30-year half-life. However, this effect has been simulated with neutron irradiation of ^{133}Cs bearing silicate and aluminate glasses and pollucite (GRAY 1982).

Naturally occurring ^{133}Cs has a fairly large thermal neutron capture cross-section (29 barns) and can be converted to ^{134}Cs which decays to Ba with a 2.1-year half life. Unfortunately, ^{134}Cs also has a fairly large neutron capture cross-section and yields the very long-lived ^{135}Cs (half life $> 10^5$ y) isotope. Therefore, the amount of ^{134}Cs that can be stabilized with neutron irradiation is limited, and does not continue to increase with neutron dosage. The early work by Gray (1982) showed that only 12% of the ^{133}Cs was converted to ^{134}Cs by neutron capture. In addition, irradiated samples needed to be annealed to remove neutron damage effects that can mask changes caused by subsequent transmutation. Two years following irradiation, approximately 6% of the total Cs had undergone transmutation or only 13% of the transmutations expected in a actual waste sample. Nevertheless, small density and leachability increases were observed for the silicate glass and pollucite samples. X-ray diffraction studies of the pollucite samples suggested that they were at least partially crystalline (GRAY 1982).

History of the ^{137}Cs Pollucite Samples

In the early 1990's, the French company, Commissariat A L'Energie Atomique Office des Rayonnements Ionisants (ORIS), inquired about the purchase of a capsule of CsCl from

Westinghouse Hanford Company. These capsules of CsCl were produced at Hanford from the early 1960's to the 1970's in an effort to reduce the heat load on the underground storage tanks. These capsules are stored at Hanford in the Waste Encapsulation and Storage Facility. Occasionally, capsules of CsCl or SrF₂ were sold or loaned as irradiation sources or as sources of ¹³⁷Cs or ⁹⁰Sr. In this case, ¹³⁷Cs was needed at ORIS to make tumor irradiation sources. In trade for a large capsule of CsCl, a suite of 40 small irradiation capsules was requested and received. These capsules have been stored at Pacific Northwest National Laboratory since being received from ORIS.

Originally fabricated as radiation sources for tumor treatments, the samples are sealed, 300 micrometer-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; there are six samples in each age range. An additional six samples are available in which no ¹³⁷Cs was incorporated. Baseline data will be collected on these nonradioactive samples.

Capsule and Pollucite Characterization - J. A. Fortner

The first of the sample capsules containing a nonradioactive pollucite was opened by cutting the welded end with a Buhler Isomet[®] saw. The sample was found to be a small pressed-powder pellet, approximately 4-mm diameter by 1-mm thickness, which retained its shape upon removal from the capsule (Figures 1-3). Because detailed drawings of the capsules were unavailable, we had to cut the stainless steel capsule in a stepwise fashion, making measurements of the dimensions as it was opened. The capsule was found to have a uniform wall thickness of 0.5 ± 0.05 mm, and the sample was located at the machined bottom, not readily accessible from the welded top of the capsule. This information is important when we begin opening the capsule's housing actual radioactive samples, some of which have surface dose rates of 0.2 Gy (20 rad).

Samples of pollucite powder from the opened capsule were examined in an analytical electron microscope (AEM). The first samples of material to be examined in the AEM were embedded in Medcast[®] epoxy, which did not produce satisfactory sections with the pollucite. A different epoxy, LR White, was subsequently used for the embedding prior to microtoming. This epoxy provided superior sections to the previous attempt, where most of the sample was torn from the sections. The pollucite was found to be somewhat beam-sensitive in the 200 keV AEM, probably due to a knock-on effect from hydration of the sample (Figure 4). Some of the material was re-examined after being desiccated in the microscope for -48 hours, followed by liquid nitrogen cooling (Figure 5). The pollucite was much more resistant to radiation damage when treated in this manner. In addition to the pollucite, energy dispersive X-ray fluorescence spectroscopy (EDS) revealed a minor amount of CsAlSiO₄ phase (Figure 6). Previous X-ray analysis placed this material as -10% of the sample by weight. It appeared to be more refractory than the pollucite, appearing as a thick chunk (that 200 keV electrons would not penetrate) within the section rather than as a thin-sectioned component of the sample. Also employed was electron energy loss spectroscopy (EELS), a technique analogous to near-edge X-ray analysis of structure (NEXAS), from the oxygen K-edges, aluminum and silicon L-edges, and cesium M-edges (Figures 7 and 8). These edges are expected to be sensitive to focal atomic structure; it will be instructive to compare these results from microscopic samples with X-ray results from the bulk specimens. Some evidence for electron beam damage affecting the aluminum near edge structure can be observed in Figure 8. The silicon edge will be less sensitive to structural changes, because of the stability of the SiO₄⁴⁻ tetrahedral structure.

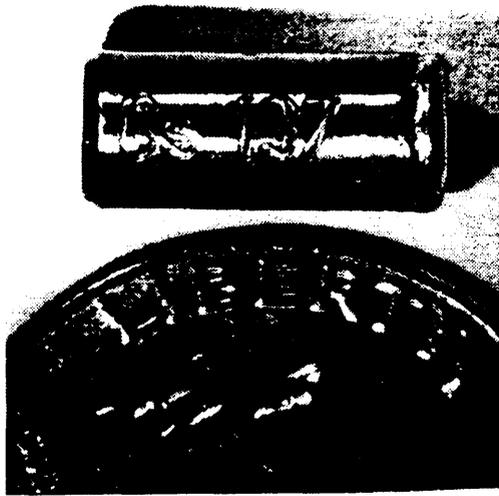


Figure 1. Pollucite capsule, with dime, for scale. This capsule contained only nonradioactive cesium, despite the stamp.

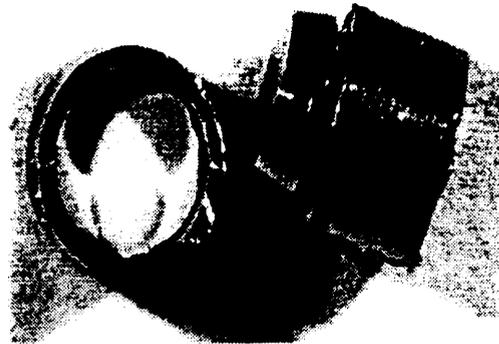


Figure 2. Opened capsule containing nonradioactive pollucite. When opened from the welded top, this capsule was found to contain a powder that appeared to have been pressed *in situ* to the capsule. A stainless steel disk was found just beneath the welded closure and above the pressed powder pellet.



Figure 3. Close-up of the pressed pollucite pellet in the opened capsule. The pellet is approximately 1.5-mm dia by 4-mm thick and rests on the non-welded end of the capsule (0.5-mm thick).

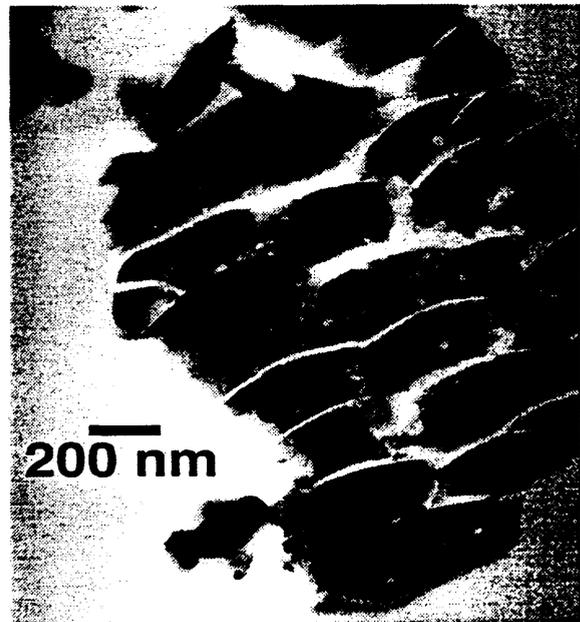


Figure 4. The AEM micrograph of pollucite from the opened capsule. Arrow indicates knock-on radiation damage symptomatic of hydration.

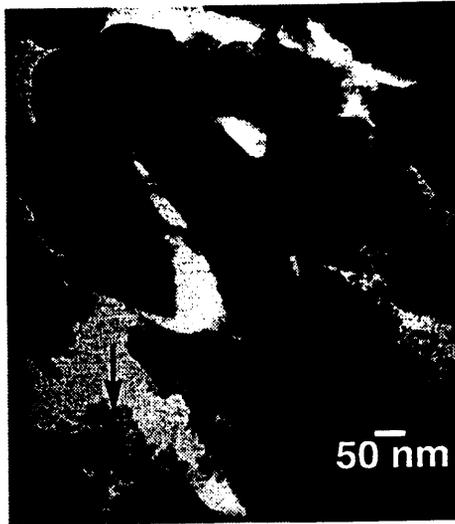


Figure 5. The AEM micrograph of pollucite from the capsule after the microtomed sample was allowed to desiccate in the microscope over the weekend. The arrow indicates a portion having a distinct morphology, containing excess Al and low Cs.

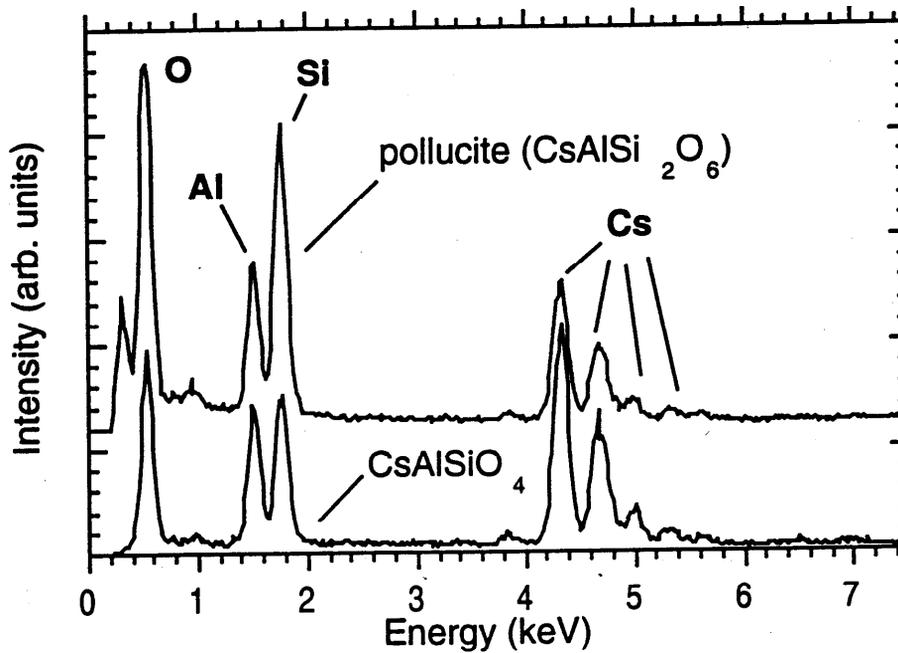


Figure 6. Energy dispersive X-ray fluorescence spectra showing presence of pollucite (top curve) and CsAlSiO₄ minor phase (bottom curve). -The spectra were obtained in the AEM on samples from the opened capsule.

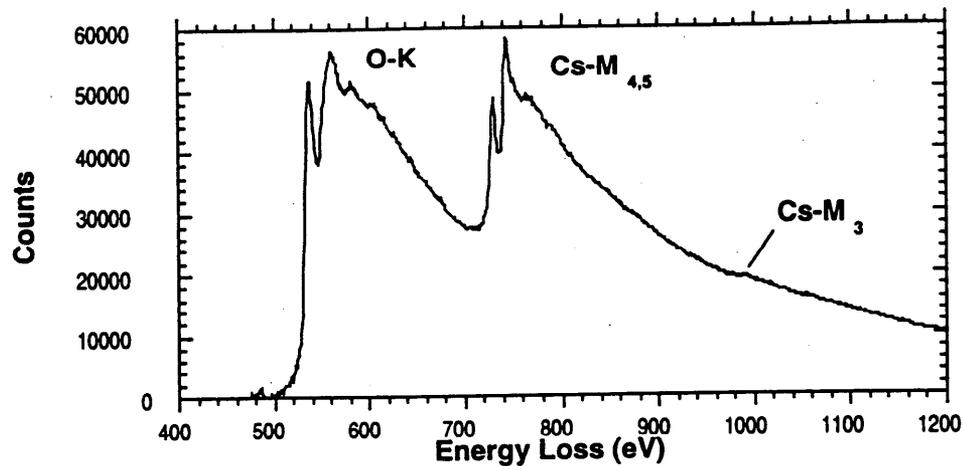


Figure 7. Electron energy loss spectrum of oxygen K-edge and Cs M-edge of pollucite from the nonradioactive capsule. The oxygen edge shows a fair amount of near-edge structure, which is generally sensitive to changes in the chemical environment.

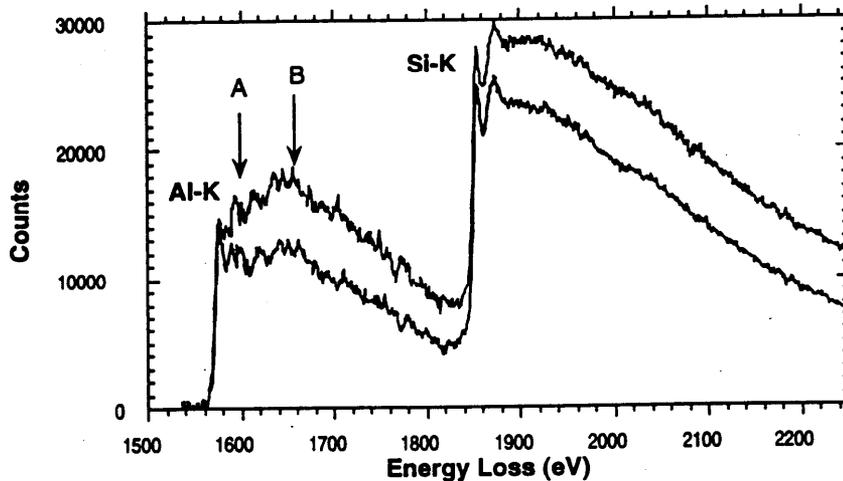


Figure 8. Electron energy loss spectra of Al and Si K-edges in undamaged (lower spectrum) and electron-beam damaged (upper spectrum) pollucite from the nonradioactive sample. The peak structure of Si is unaffected by the electron damage, while the Al edge shows changes in near edge structure (A) and in collective electronic contributions (plasmons, B).

Electron Radiation Effects in Pollucite - Edgar C. Buck

Pollucite, ideally $\text{CsAlSi}_2\text{O}_6 \cdot 0.5\text{H}_2\text{O}$, is a framework silicate phase, related to analcime. The phase is cubic with $a = 1.3673$ nm, although reported values vary between 1.366 and 1.374 nm. Pseudo-cubic structures have also been reported, which have a variable water content. Natural pollucite is found in highly fractionated peraluminous pegmatites, where it is frequently penetrated by several generations of feldspar-, mica- and quartz-bearing veins, replaced by (K, Rb)-feldspar, and subsequently argillized (TEERTSTRA et al. 1995). The Si/Al ratios in pollucite range from 2.4 to 2.6. Hydrothermal fluids can cause the generation of (K, Rb)-feldspar and analcime by cation exchange along fractures and vein surfaces. However, a fine-grained white solid can be prepared through synthetic routes.

Experimental Procedure

After the steel capsule was cut open, material from the center of the pressed pellet was placed on an aluminum stub, carbon coated, and examined with scanning electron microscopy (SEM). This examination, combined with EDS compositional analysis confirmed that the capsule contained pollucite; however, on close inspection, it was evident that another Cs-bearing phase was present. This phase was a Cs-containing silicate. The morphology and distribution of this phase suggested that it may be binding the pollucite particles together in the capsule.

Small particles of the pollucite were taken from the steel capsule and placed into polyethylene capsules which had been filled with a few milliliters of LR White acrylic. After curing for 24 hours, the sample block was trimmed and thin-sectioned with an ultramicrotome. The electron transparent sections were examined with an AEM. The composition of the phases was determined with EDS and EELS. This method of sample preparation allowed large areas of pollucite to be prepared, which greatly assisted in obtaining suitable regions for electron irradiation studies. Examination with was selected because we had successfully used it, in an earlier study, to probe the composition and structure of small uranium phases within a matrix of other components in contaminated soil. Quantification of the EDS analysis was performed by use of experimentally determined k-factors obtained from glass and mineral thin film standards. Phases were identified by a combination of EDS and electron diffraction. The AEM camera was calibrated by use of a polycrystalline aluminum sample. Electron diffraction patterns for d-spacing estimation were made while a steadfast eucentric height and objective lens current were maintained and the brightness control defocused.

Results of Electron Beam Irradiation

Samples of pollucite were irradiated for up to 1 hour with an intense 200 keV electron beam in the AEM. The crystals of pollucite were oriented as close as possible to a major zone axis. During these irradiations, the damage process was monitored by use of electron diffraction patterns and AEM images, along with EDS and EELS. As expected, irradiation resulted in the volatilization of Cs from the pollucite phase. In Figure 9, the loss of Cs can be seen during an irradiation; however, further loss of Cs after 10 min was not observed.

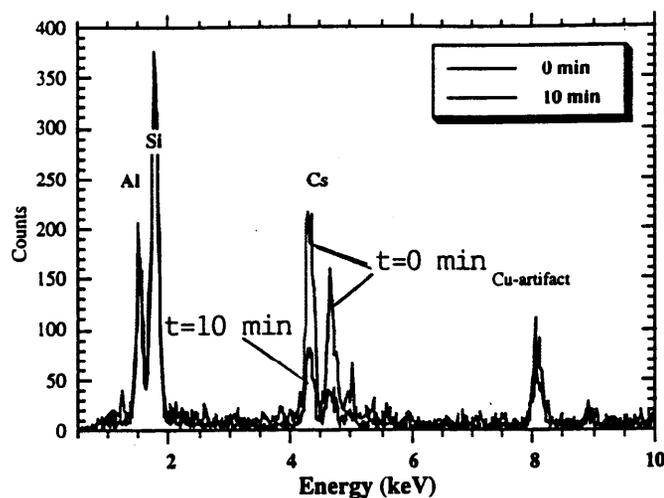


Figure 9. Plot showing the change in composition during electron irradiation in pollucite. Over a 10-minute irradiation, the cesium concentration dropped over 50%. Note that the spectra have been normalized to Si.

Atomic displacements in pollucite may be either produced by knock-on displacement or by a combination of knock-on displacement and radiolysis. The displacement probabilities may be different for each atom type in the phase. The displacement energies (E_d) will depend on bond strength, the space available for accommodating an interstitial, and the form of the interstitial. Displacement events are high energy events that are characterized by a threshold energy, below which the event will not occur no matter how intense the electron flux. With a 200 keV beam it can be difficult to separate the possible effects of heating (melting) from displacement damage which results in amorphization. We do not expect that the material melted in the electron beam, since the melting point of pollucite is $>1700^\circ\text{C}$, and it is unlikely that a temperature even close to this value was reached during the irradiation. Evidence from the AEM study suggested that melting and recrystallization did not occur; therefore, we concluded that we had observed displacement damage events.

Figure 10, a series of electron diffraction patterns taken along the [01 1] zone axis, tracks the damage to the pollucite during an irradiation. After 4 minutes (Figure 10a to c), the sample appears to remain crystalline. Note that the intensity of some spots changes within this time period. This may have been caused by slight movement in the crystal, possibly caused by heating from the electron beam. In Figures 10c and f, it is evident that the material is now nearly completely amorphous. The diffuse halos are more prominent; however, a few diffraction spots remain.

Figure 11 and 12 indicate that there is a rapid loss of crystal structure, possibly the result of minor disorder; however, the material remains crystalline and the composition does not change. The diffraction patterns indicate that the intensity of reflections decreases, but the reflections remain. This may be a heating effect (i.e., Debye-Waller factor). At irradiation times longer than 10 min, Cs loss occurs, and the material starts to become amorphous.

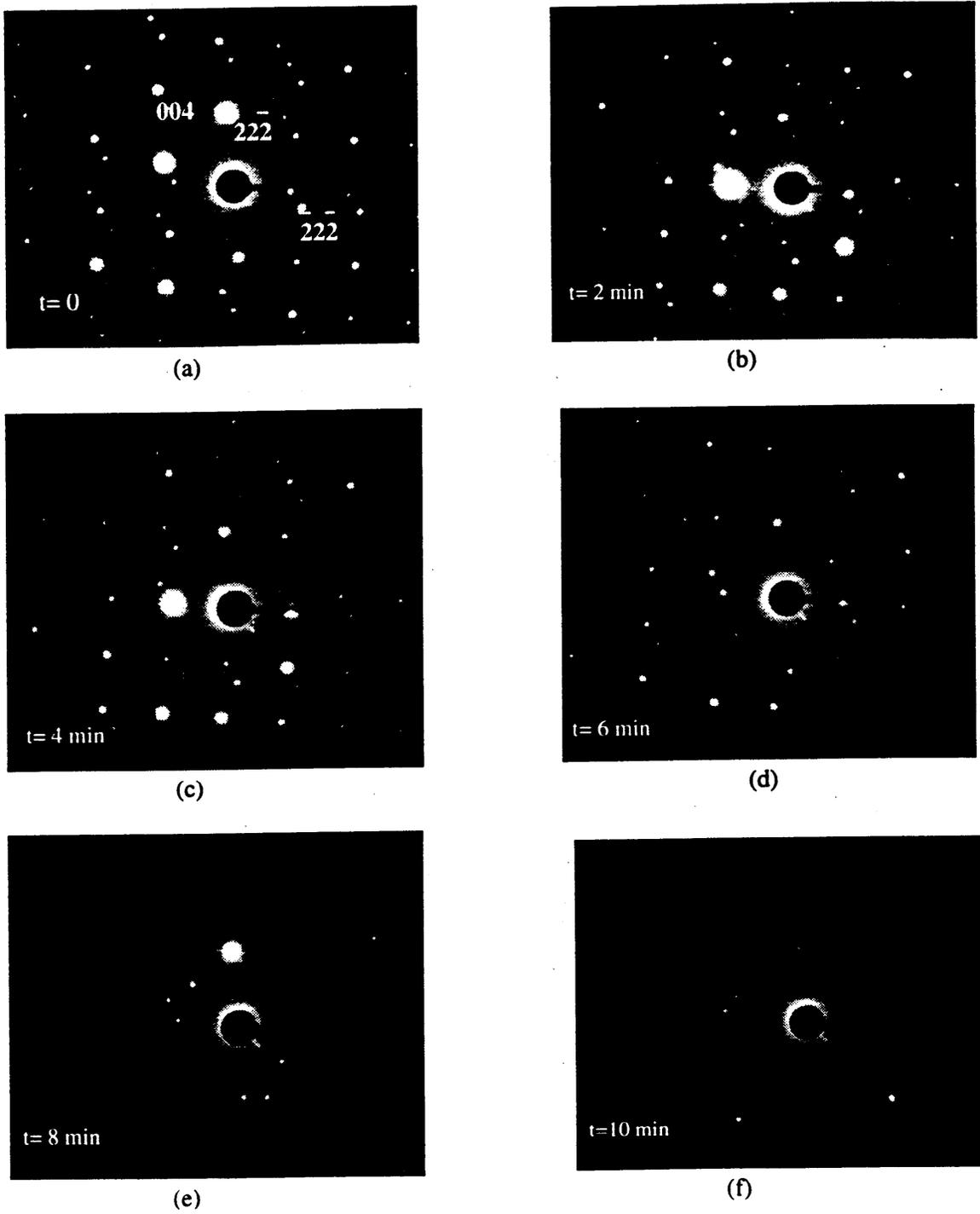


Figure 10. Electron diffraction from a pollucite sample at various levels of electron irradiation: (a) 0 min, (b) 2 min, (c) 4 min, (d) 6 min, (e) 8 min, and (f) 10 min.

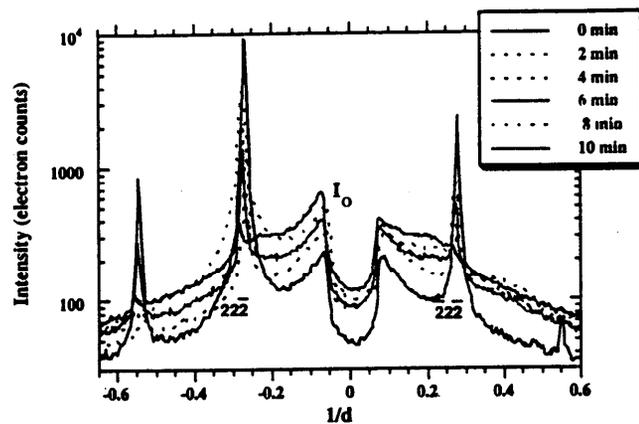


Figure 11. Electron diffraction profile of the $\langle 11 \rangle$ systematic row in pollucite. The diffraction signal at $t=0$ min is the strongest and rapidly degrades with irradiation time; however, a diffuse background steadily increases with the irradiation time. This effect is particularly strong around the central peak.

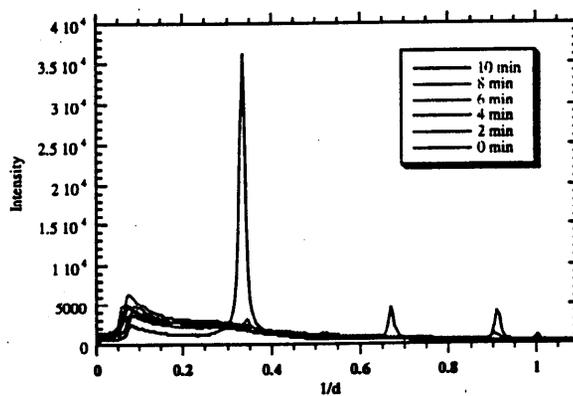


Figure 12. Another electron diffraction profile showing the decrease in intensity of the diffraction peaks during an irradiation. The data have been extracted from a minor systematic row.

During electron irradiation in the transmission microscope, there can be mutual annihilation of vacancies and interstitials, annihilation at the thin-section surface, interactions with impurity atoms, and absorption at internal sinks such as grain boundaries **and** dislocations. Agglomeration of the same type of point defects may produce dislocation loops, stacking fault tetrahedra, and voids. These types of structures, however, have not been observed in the present set of irradiations. At the beginning of an irradiation, interstitials will tend to either be annihilated at vacancies or be trapped at another interstitial, resulting in the nucleation of a dislocation loop. Toward the end of an irradiation, dislocation loops will grow, as interstitials will tend to add to existing loops rather than nucleate new ones (BROWN et al. 1969). Because evaluated irradiation temperatures cause interstitial loops to nucleate and grow rapidly, we will be conducting further irradiations at elevated temperatures.

By noting the voltage when the first small clusters produced by irradiation are visible in the electron microscope, the minimum accelerating voltage can be calculated by extrapolation. This value can then be used to calculate E_d . However, this measurement requires the use of an electron microscope which can irradiate the sample at much higher energies than have been used in this initial study. The main advantages of the AEM are that the electron energy can be controlled, and that direct observation of the growth of dislocation loops and other phenomena is possible. The microscope also allows control of crystallographic alignment, as demonstrated in this work, and there is the possibility of temperature control. Because a crystal lattice is not isotropic, the energy required to displace an atom from a lattice site will depend on the direction in which the atom moves; this effect is more pronounced in an anisotropic lattice such as calcium titanate, but even in the cubic lattice of pollucite, there may be crystal directions that are more susceptible to irradiation than others.

The electrons that impinged on the samples during these irradiations represent, even at the shortest irradiation time, many thousands of years of exposure to the electrons from the normal β decay. Of course, the ^{137}Cs with a half-life of 30 y would have already decayed by the time any significant radiation damage would have occurred. These results suggest that crystallinity of the radioactive pollucite is likely to be unaffected by the β radiation and any damage that is noted may be a result of the transmutation effects.

X-Ray Absorption Spectroscopy of Sealed Sources - N. J. Hess

The primary goal of the XAS experiments on the sealed sources is to evaluate the stability of the pollucite, the host ceramic material. The stability of host ceramic is in question because the transmutation of ^{137}Cs to ^{137}Ba results in the accumulation of Ba atoms in the pollucite structure. The solubility of Ba within pollucite is known to be about 10 wt% (LANGLET 1969). However, the fate of the pollucite structure at Ba concentrations greater than the solubility limit as a result of transmutation is not known. It is possible that a second, Ba-rich oxide phase forms, or that metallic Ba clusters are created. In the later case, air exposure of aged pollucite material containing small, metallic Ba clusters could result in a highly exothermic reaction. These initial XAS experiments are designed to reveal the presence of metallic Ba clusters. If present, these metallic clusters will be apparent in both the Ba XANES and EXAFS. The absence of metallic Ba would allow removal of the pollucite ceramic material from the sealed sources. This would greatly expand the assortment of experimental probes that can be brought to bear on determining the effects of Cs transmutation on the pollucite structure.

A custom sample holder for the sealed sources has been designed and fabricated to provide shielding from the gamma radiation while maximizing the solid angle that the fluorescence signal can be collected. The sample holder mates to an existing liquid nitrogen cryostat cold finger with

a dovetail assembly that minimizes the time required to mount the samples yet provides good thermal contact with the cold finger.

In preparation for X-ray absorption spectroscopy (XAS) experiments on the sealed sources, standard Cs and Ba samples were analyzed at the Stanford Synchrotron Radiation Laboratory during January and March of 1997 on a "wiggler" side station at the Cs and Ba K and L_{III} absorption edges. Analyzed in addition to the Cs and Ba standards were Ba-doped samples of pollucite, CsAlSi₂O₆, and the Ti analog, CsTiSi₂O_{6.5}. The XAS data were collected in fluorescence mode using a 13-element Ge detector. In the fluorescence signal from samples that contained both Cs and Ba, the Cs fluorescence emission could not be separated from that originating from Ba at the La-edge. However, at the K-edge, the Cs and Ba fluorescence emission was well separated because of the enhanced energy discrimination capabilities of the customized amplifiers. Analysis of the XAS data from the Cs standards and the Ba-doped pollucite samples demonstrated that a Ba X-ray absorption near edge structure (XANES) can be obtained on samples containing as little as 0.25 mass % Ba. Unfortunately, analysis of the extended X-ray absorption fine structure (EXAFS) spectra of the Ba-doped samples was limited due to severe problems with the monochromator crystal.

The XAS experiments planned for June 1997 will be conducted on a wiggler end station, where there will be nearly an order of magnitude increase in flux over the side station used in March 1997. The increased flux will increase the intensity of the fluorescence signal and decrease the amount of Ba required for collection of Ba XANES

The 40 ¹³⁷Cs sources received from ORIS were manufactured over the period 1972 to 1989 for tumor treatment and contain varying amounts of the ¹³⁷Cs isotope. Since the half-life of ¹³⁷Cs is approximately 30 years, the sources have accumulated a wide range of Ba atoms as a result of the transmutation of Cs to Ba. The ¹³⁷Cs sources were retrieved from storage, and for the first set of XAS experiments, six sealed sources were selected for analysis. These six initially contained between 1 and 24 wt% ¹³⁷Cs and currently contain between 0.1 and 7 wt% Ba. They will be prepared for the XAS experiments by thinning one end of the stainless steel capsule. Thinning the steel of the capsule was shown to be necessary from the XAS experiments conducted in March 1997 on identical sealed sources that did not contain any ¹³⁷Cs isotope. The signal-to-noise ratio of the fluorescence signal from these cold sources was not sufficient to allow analysis by EXAFS. Thinning the stainless steel bottom of the capsule from 0.5 mm to 0.125 mm will increase the fluorescence signal by one order of magnitude. Further gains in the fluorescence signal will be realized by cooling the sources to near liquid nitrogen temperatures and by employing the wiggler end station at the Stanford Synchrotron Radiation Laboratory. To thin the capsules with precision, a small lathe has been purchased. After the capsule bottom has been thinned, the exterior of the capsule will be checked for ¹³⁷Cs contamination and checked again after being quenched to liquid nitrogen to assure that the integrity of the encapsulation has not been compromised.

Future Work

The work carried out this year has been in preparation for examining the radioactive pollucite samples. Examination of the radioactive samples will begin at the Stanford synchrotron once the equipment modifications have been made and the safety plans written. Determination of the state of the Ba in

1) A term used to describe how the beam is directed where some X-ray intensity is utilized at side and end stations. The intensity of the beam at the side-stations is less than the intensity at the end of the beam line.

pollucite may provide some relief in how carefully the remaining studies need to be carried out; for example, after the capsule has been opened, examinations of the pollucite under inert atmosphere may not be needed. Heavy ion irradiations will be performed in the near future with the pollucite samples. In the case of heavy ion irradiations, isolated Frenkel pairs form along the path of the incoming particle. These irradiations often result in an overall volume expansion if there is no clustering of defects.

Examination of the nonradioactive Ba-doped pollucite and Ti pollucite analogues will also continue so that a baseline is obtained for the XAS work. In this regard, the examination of the Ba-doped and Ti pollucite analogue and the pollucite samples from the capsules gives information that supports the interpretation of the XAS work. We will also explore the effect of electron irradiation on barium-doped samples of pollucite.

References

- ANCHELL, J., WHITE, J, THOMPSON, M. R., AND A. C. HESS. 1993. "Ab initio periodic Hartree-Fock study of Group 1A cations in ANA type zeolites" *Journal of Physical Chemistry*.
- BROWN, L. H., KELLY, A., and MAYER, R. M. (1969) *Philos. Mag.* **A19, 721**.
- CERNY, P., and LENTON, P. G. (1995) The Buck and Pegli Lithium Deposits, Southeastern Manitoba --The Problem of Updip Fractionation in Subhorizontal Pegmatite Sheets, *Eco. Geol. and the Bull. of the Soc. Eco. Geol.* **90,658-675**.
- GRAY, W. J. 1982. "Fission product transmutation effects on high-level radioactive waste forms." *Nature*, 296(5857):547-549.
- HOGAN, M. A. and RISUD, S. H 1991. "Gel-derived amorphous cesium-aluminosilicate powders useful for formation of pollucite glass-ceramics." *Journal Materials Research*, 6(2):217-291.
- KLAVATTER, E. A., BROWN, N. E., TRUSDELL, D., ANTHONY, R. G., GU, D., and THISBAUD-ERKEY, C., (1994). "Ion-Exchange Performance of Crystalline Silicotitanates for Cesium Removal from Hanford Tank Waste Simulants." In Proceedings of Waste Management '94, University of Arizona, Tucson, AZ.
- LANGLET, G. (1969). "Synthese et Etude Cristallographique des Composes du Systeme Cs₂O-Al₂O₃-SiO₂." Technical Report No. CEA-R-3853, Commissariat A L'Energie Atomique, France.
- STRACHAN, D. M. and SCHULZ, W.W. (1979). "Characterization of pollucite as a material for long-term storage of Cesium- 137." *American Ceramic Society Bulletin*, 58(9):865-869.
- TEERTSTRA, D. K., CERNY, P., and NOVAK, M. (1995) Composition and Textural Evolution of Pollucite in Pegmatites of the Moldanubian, *Miner. Petrol.* **55,37-55**.