Spatially resolved optical absorption spectrometry and single crystal diffraction on metamict materials

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A major goal in developing storage medium for radioactive waste is the identification of chemically suitable and durable material for storage in repositories (Lumpkin 2006). Radiation damage induces enhanced chemical diffusion and structural breakdown of the host materials, which can lead to contamination of the surrounding environment. During this project four different naturally occurring materials which are common carriers of thorium and uranium were examined : gadolinite, perrierite, allanite, and pyrochlore of which the first three are silicates and pyrochlore being an oxide. Their spectra and absorptions bands were examined to identify prominent features due to radiation damage.
The goal of this study is to identify and characterize polyamorphisms metamict glasses. Further, we examine the hypothesis that pyrochlores do not amorphise but undergo a structural transition upon metamictization this part of the project will be conducted at the APS.

Spatially Resolved Optical Absorption Spectrometry And Single Crystal Diffraction On Radiation-Damaged Materials.

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Introduction:

A major goal in developing storage medium for radioactive waste is the identification of chemically suitable and durable material for storage in repositories (Lumpkin 2006). Radiation damage induces enhanced chemical diffusion and structural breakdown of the host materials which can lead to contamination of the surrounding environment. During this project four different naturally occurring materials which are common carriers of thorium and uranium were examined : gadolinite, perrierite, allanite, and pyrochlore of which the first three are silicates and pyrochlore being an oxide. Their spectra and absorptions bands were examined to identify prominent features due to radiation damage.

Gadolinite; Ytterby; Iveland	$Y_2FeBe_2Si_2O_{10}$
Width: 40 µm	Height: 108 µm
Perrierite, Amherst VA	$(Ce,La,Ca)_4(Fe^{2+},Mg)_2(Ti,Fe^{3+})_3Si_4O_{22}$
Width: 2 µm	Height: 95 µm
Allanite	$Ca(Ce,La,Y,Ca)Al_2(Fe^{2+},Fe^{3+})(SiO_4)(Si_2O_7)O(OH)$
Width: 19 µm	Height: 89 µm
Pyrochlore	$(Na,Ca)_2Nb_2O_6(OH,F)$

Varied in thickness and diameter between 30-50 µm

(Mineral Database 2006)

Spectroscopy and Absorption Techniques:

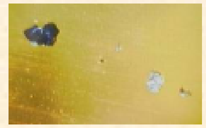
Being relatively large, the samples were mounted onto capton tape for spectroscopy. Two sets of data were collected at once: one being the spectrum of the sample and the other a spectrum of the white light source without interference.When analyzing the data, the white light spectrum was subtracted from the spectra of the samples (Figure 3) as well as the capton spectrum being divided into the sample spectra to provide the final normalized intensity (Figure 4).

Spectroscopy and Absorption Analysis:

Gadolinite have prominent dark regions against the green transparent overall color. The dark regions indicate areas that suffer radiation damage. The two spectra are layered to view any differences (Figure 1). When the spectra are subtracted from each other after being normalized, the varying features, if any, can be seen (Figure 2).

The three silicate samples ranged in degrees of radiation damage. Perrierite and gadolinite having high and allanite having a low degree of damage. All the spectra have dominant features in the visual band of the spectrum which is caused by the Fe3+ and Fe2+ charge transfer. The dominant absorption feature is also the Fe2+ and Fe3+ charge transfer band (Sherman 1987).

Spectroscopy Instrument, UNLV Physics Lab 139



Gadolinite, Perrierite, and Allanite on capton

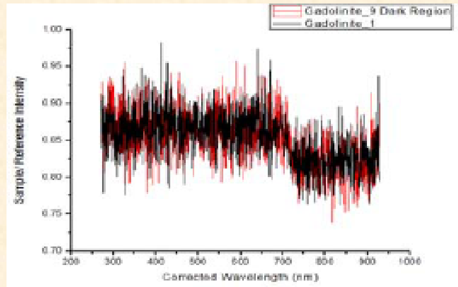


Figure 1

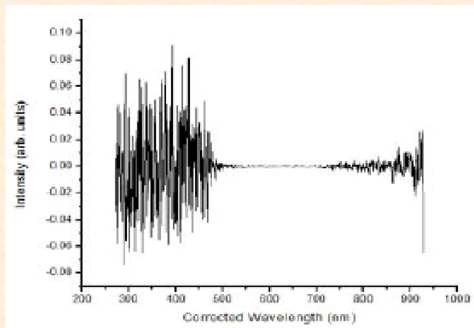


Figure 2

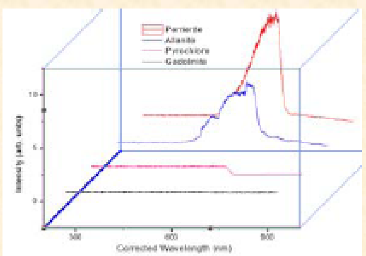


Figure 3

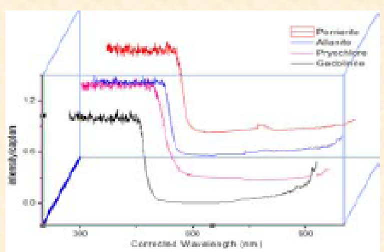


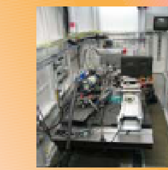
Figure 4

Examination with X-Ray diffraction:

Morphological single crystals of pyrochlore were also examined using an X-ray diffraction instrument, 16BM-D, at Argonne National Lab.

Some of the pyrochlore crystals had recrystallized, which is known as powder (Figure 5).

Where as others were still crystals with varying degrees of modest radiation damage (Figure 6).



16BM-D at Argonne National Lab, Photo Credit: www.anl.gov

Figure 5

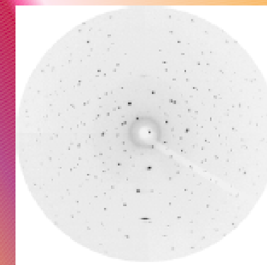


Figure 6

Conclusions:

The spectra for all four samples, as well as the absorption bands, are dominated by Fe-charge transfer band which is expected due to Fe content in the samples. However, the optical absorption spectra do not have any defining characteristic features of radiation damage nor is there any noticeable difference between the damaged and undamaged gadolinite regions so consequently there is also no spatial variation even though the composition of gadolinite is varied.

The pyrochlore samples, fifteen in total, were also dominated by Fe in the optical absorption spectra. Only two of the fifteen samples of pyrochlore had crystal lattices, as shown in Figure 6, where as the rest of the samples were powder.

References:

1.David Barthelmy, Mineralogy Database, http://www.webmineral.com.

2.Lumpkin, Gregory R. 2006. Ceramic Waste Forms for Actinides. *Elements* 2006, vol. 2: 365.

3.Sherman, David M. 1987. Molecular Orbital (SCF-X α -SW) Theory of Metal-Metal Charge Transfer Processes in Minerals. *Physics and Chemistry of Minerals 14*, 364.

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Programs:

Origins 8.0, Winspec, PowerPoint, Excel