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

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

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.
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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 (Lumadue 2005). Radiation damage induces enhanced chemical diffusion and structural breakdown of the host materials which can lead to contamination of the surrounding environment. During this process four different naturally occurring minerals which are common carriers of thorium and uranium were examined: gadolinite, parnellite, atlantite, and pyrochlore of which the first three are silicates and pyrochlore being an oxide. Their spectra and absorption bands were examined to identify prominent features due to radiation damage.
Gadolinite, Yttria: Ireland
Y2Fe4+Fe3+Si2O7
Width: 43 μm Height: 108 μm
Parnellite, Amphorst VA
(Ca,La,Ce)2(Fe2+,Mg)2(Ti,Fe3+)2SiO2
Width: 2 μm Height: 96 μm
Atlantite
Ca(Ca,La,Y, Ce)2(Fe2+,Fe3+)3SiO6(O,Si)2O(OH)
Width: 10 μm Height: 99 μm
Pyrochlore
(Na,Ca)2Zr2SiO7(OH,F)
Varied in thickness and diameter between 30-50 μm
(Minerals Database 2006)

Spectroscopy and Absorption Techniques:
Being relatively large, the samples were mounted onto kapton tape for spectroscopy. Two sets of cuts 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 kapton 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. The grey 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 will be seen (Figure 2).
The three silicate samples ranged in degrees of radiation damage. Parnellite and gadolinite having high and atlantite having a low degree of damage. All the spectra have prominent 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 Fe3+ and Fe2+ charge transfer band (Sherman 1987).

Examination with X-Ray diffraction:
Morphological single crystals of pyrochlore were also examined using an X-ray diffraction instrument, 10BM-D, at Argonne National Lab.
Some of the pyrochlore crystals had recrystallized, which is known as powder (Figure 5).
Whereas others were still crystals with varying degrees of modest radiation damage (Figure 6).

Conclusions:
The spectra for all four samples, as well as the absorption bands, are dominated by Fe2+ 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 Fe2+ in the optical absorption spectra. Only two of the thin samples of pyrochlore had crystal lattices, as shown in Figure 6, whereas the rest of the samples were powder.

References:

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