Microbeam Studies of Cs Adsorption by Hanford Micas

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A major component of Hanford waste is radioactive ^{137}Cs. Previous study has determined that the Cs is preferentially adsorbed by micaceous minerals in the soil. While it is possible in some cases to image the Cs in the electron microscope, the x-ray microbeam at the PNC-CAT combined with fluorescence detection can provide greater sensitivity to low levels of Cs and also provides spectroscopic information.

Mica grains (biotite and muscovite) were extracted from Hanford soils and exposed to dilute Cs solutions. The samples were then imaged using Kirkpatrick-Baez mirrors to provide microbeams ranging from 1-5 mm. Typically, the adsorbed Cs is concentrated in small regions at the edges or interior cracks of the samples (see Fig. 1).

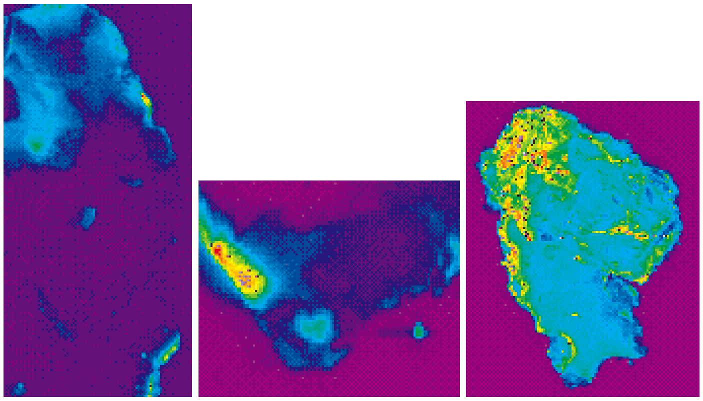

FIG. 1. Some Cs images from three biotite samples. Left: image size 270 × 620 microns; Middle: image size 140 × 100 microns; Right: image size 950 × 1200 microns. The bright red and yellow regions are high Cs concentrations.

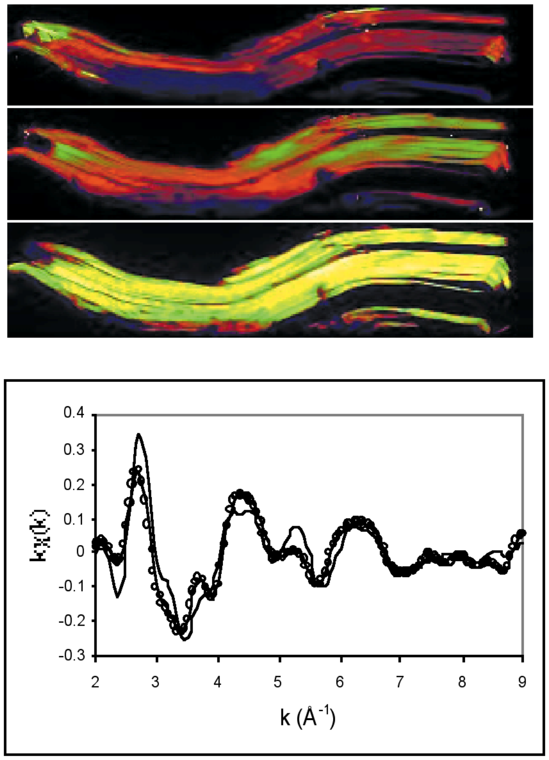

FIG. 2. Top: X-ray microprobe images of a sectioned biotite particle. The image sizes are 780 × 165 microns with a beam size of 1 micron. The top image is the Cs map, the middle is for Mn, and the bottom for K. Highest concentrations are indicated by yellow and lowest by blue or black. Each plot is scaled to its maximum concentration. The maximum Cs concentration is much less than for the other elements. Bottom: Mn XAFS from a high Cs (points) and a low Cs (line) area.

To look more closely at this process, a series of mica grains were sectioned transverse to their basal planes prior to exposure to Cs. As shown in Fig. 2, the Cs again concentrated in small regions, indicating that the preference for edge sites is not due to kinetic constraints of Cs transport into the interior of the mica grains. Rather it is likely some type of weathering process makes certain regions more receptive to Cs adsorption. As Fig. 2 indicates, many elements can be simultaneously imaged with the solid-state detector. Typically, the concentrations of Fe, Mn, Ti, and K were determined along with Cs. For this sample, the Cs-rich region correlates with a low concentration of K. Chemically, it can be expected that the Cs will replace K in the mica. However, the K depletion is much larger than the adsorbed Cs.

Using the microprobe, it is also possible to obtain spectroscopic information. To look for more information on the Cs-rich site, x-ray absorption fine structure (XAFS) spectra were obtained for Cs, Mn, Ti, and Fe, and compared to similar spectra from a region with low Cs adsorption. Surprisingly the spectra were nearly identical in the two regions. The only difference was in the Mn XAFS (see Fig. 2), although even in this case the changes were minor. Unfortunately, as currently configured it was not possible to obtain the K XAFS spectrum on the ID beamline. This shortcoming is not an intrinsic limit and will be remedied in the future.

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