Paramagnetic Defect Centers in Hydrothermal Kaolinite from an Altered Tuff in the Nopal Uranium Deposit, Chihuahua, Mexico

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Abstract: Point defect centers in hydrothermal kaolinite have been investigated using electron paramagnetic resonance (EPR). Kaolinite was sampled in petrographically well-defined materials coming from uranium-rich hydrothermally altered volcanic tufts (Nopal I uranium deposit, Chihuahua, Mexico), which show extensive kaolinization and an intense redistribution of uranium. Several kaolinite parageneses were defined according to their origin (fissure fillings and feldspar pseudomorphs); their location relative to the U^{6+} mineralization at the scale of the deposit (mineralized breccia pipe vs. barren surrounding rhyolitic tufts), and at the scale of mineral assemblages; and their crystal chemistry.

Two types of centers of axial symmetry were identified (A- and A′ -centers) and represent positive holes trapped on apical oxygens (Si—O^--centers). A-centers were stable to 400° C, whereas A′ -centers annealed at 350° C. A relation between defect-center concentration and U content demonstrates that natural irradiation was responsible for these centers. On the other hand, defect-center concentration was not directly linked to the origin (fissural or feldspar pseudomorph) or the crystal chemistry (structural order and substitutional Fe^{3+} content) of the kaolinite, According to petrographic data, and with respect to the relative thermal stability of A- and A′ -centers, two successive irradiations of kaolinite were evidenced: (1) originally during crystallization of kaolinite from radioactive hydrothermal solutions, and (2) permanently when kaolinite was in contact with secondary U-silicates, which led to the formation of A′ -centers.

Because of the short half-life of U, these two radiation-induced centers were created by short-lived elements of the U-decay series. As a consequence, variations of defect-center concentration possibly reflect variations in radioactive disequilibrium during the history of the alteration system. This provides a unique tool for tracing the dynamics of the transfer of radionuclides in the geosphere: kaolinite may be used as a sensitive *in situ* dosimeter, which may be useful in the fields of weathering petrology and nuclear waste management.

Key Words: Defect centers • Electron paramagnetic resonance • Hydrothermal • Kaolinite • Natural radiation

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