Tracing past transfers of radionuclides with radiation-induced defects in clay minerals

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Understanding the processes controlling migrations of radioelements at the Earth's surface in an important issue for the long-term safety assessment of high level nuclear waste repositories as well as for prospection. Evidence of past occurrence and transfer of radionuclides can be brought using radiation-induced defects in clay minerals. Such defects in kaolinite have been known for 35 years. They can be detected by Electron Paramagnetic Resonance spectroscopy. One defect is stable at the scale of geological periods and can be used as an tracer of radioactivity in kaolinite-containing rocks, even when radioelements have been leached away.

Based on artificial irradiations with linear accelerators or gamma sources, a methodology has been proposed to determine the paleodose cumulated by the clay mineral since its formation. The paleodose can be subsequently used to derive equivalent radioelement concentrations at the origin of the defects, provided that the age of clay minerals can be constrained. This allows quantitative reconstruction of past transfers of radioelements in natural systems. Several examples of application will be presented and discussed.

The first examples of application concern the Nopal I U-deposit (Chihuahua, Mexico), hosted in hydrothermally altered volcanic tufs, and the Coutras sedimentary deposit (France). The equivalent U-content was compared to the actual, chemically measured U-content. The results reveal past accumulations, leaching or absence of migration according to the location of samples, that in most cases are not evidenced by conventional analyses.

In recent years, radiation-induced defects have been discovered also in dickite, a polymorph of kaolinite, in smectite, in illite and in chlorite. The last example concerns defects from kaolins and illite in the proterozoic Athabasca basin (Canada). Preliminary results evidence significant migrations of radioelements since the formation of clay minerals. However, additional data on dosimetry and thermal stability are required to quantitatively interpret these results. This will widen the range of application of dosimetry with clay minerals to new parageneses and geological systems.

The role of retrograde reactions and of diffusion on ^{40}Ar-^{39}Ar mica ages

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Samples of monometamorphic metasediments from the Lepontine Alps (Switzerland) were studied, which record petrological equilibrium at 500-600 °C, 0.6-0.9 GPa. Metapelites usually contain two white mica generations, muscovite (Ms) and paragonite (Prg) or margarite (Mrg). XRD data show most mineral separates used for ^{39}Ar-^{40}Ar stepwise heating to be monomineralic. For these samples, ^{37}Ar/^{39}Ar and ^{38}Ar/^{39}Ar (i.e. Ca/K and Cl/K ratios) are constant. Ages can therefore be interpreted unambiguously. In mineral separates containing two white micas, Ca/K and Cl/K ratios are variable, reflecting non-simultaneous degassing of the two heterochemical Ar reservoirs. These ratios were used to identify each Ar reservoir and to unravel its age. Ms+Prg separates from Frodalera yield plateau ages around 16.5 Ma. In contrast, step ages are variable for a Ms+Prg separate from Molare (600 °C, 0.8-0.9 GPa), and correlate with the Ca/K ratio. The age extrapolated to the measured Ca/K ratio of Prg is 6 Ma younger than for Ms, 19 vs. 13 Ma. This is due to plagioclase (Pl) growth during decompression by partial breakdown of Prg that led to resetting of the Ar clock in Prg. In a chlorite-Mrg-biotite (Bt) calc-schist from Lucomagno (560 °C, 0.65 GPa), Bt, Mrg, and Ms all yield ages around 18 Ma. Bt separates from higher-grade rocks yield ages younger than Ms by 1-2 Ma. Minor retrograde chloritization of Bt is evident in most studied samples and is observed to be a more effective parameter than temperature in resetting the Ar clock, as is the formation of Pl from Prg decomposition. Reliable metamorphic ages require not only constraints on the PT-evolution of dated samples, but also on their microtextures.

Allanite U-Pb ages date the prograde conditions near 400 °C at 30 Ma in the northern part of the Lepontine Alps. Peak T following slight decompression was reached at 19-18 Ma, constrained by monazite U-Pb ages. Fission tracks indicate an average 'cooling rate' of 30 °C/Ma during the last 19 Ma. If so and providing that no retrogression reaction has affected the system, Ar retention in Ms is complete below ca. 520 - 500 °C, and Bt below ca. 490 °C. Retrogressed Bt and Prg can lose Ar as low as 400 °C. As at ca. 30 Ma this part of the Alpine belt was heating up, our results require that the K-Ar and Rb-Sr mica ages between 38 and 18 Ma in polymetamorphic gneisses are due to Sr and Ar inheritance and not to 'cooling' after a metamorphic peak of uniform 38 Ma age in the Central Alps.