Effects of heating on the leaching of U-Th series radionuclides in a suite of natural minerals

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The differences in the extent of leaching (with 0.1 N HNO_3 for ~25 days) and recoil of U-Th series radionuclides [U (^{234}U), Th (^{230,234}Th), Ra (^{224,226,228}Ra), Pb (^{210,212}Pb), ^{210}Po, ^{228}Ac and ^{222}Rn] from a suite of natural minerals (zircon, monazite, thorite and cerite) were assessed at room temperature and after heating these minerals at 200°C and 600°C. Our results indicate the following: i) The concentrations of U-Th series radionuclides in the leachate depend on the amount of the accumulated radiation dose in the minerals; ii) The amount of radionuclides leached from annealed (200°C or 600°C) minerals is lower by 3 orders of magnitude compared to unannealed minerals; and iii) The differences in the daughter/parent ratios of a number of pairs in the ^{238}U and ^{232}Th series (^{210}Po/^{210}Pb, ^{228}Ac/^{228}Ra, ^{234}Th/^{238}U, ^{212}Pb/^{224}Ra, ^{224}Ra/^{228}Th and ^{222}Rn/^{226}Ra) between the heated and heated minerals range over several orders of magnitude.

This study provides insight on the location of ^{238}U and ^{232}Th and possibly their daughter products in mineral grains (in paritcular, if they are located in grain boundaries), the effects of track density on the leaching of U-Th series radionuclides from mineral grains, the effects of the internal radiation damage on the escape of radon and the leaching of other non-gaseous daughter products in the U-Th series. This study has implications on the assessment of the performance of nuclear wasteforms in groundwater environment. This study has also bearing on the mobility of U-Th series radionuclides in rocks/minerals that are in contact with erupting magma.

We will present possible mechanisms for the differences in the extent of leaching of radionuclides between the heated and unheated minerals in our presentation.

Internal Lu-Hf isotope systematics of the quenched angrite D'Orbigny and two plutonic angrites

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The long-lived radioactive decay of ^{176}Lu to ^{176}Hf potentially provides a precise and disturbance-resistant chronometer for dating early Solar System processes. However, some meteorites (e.g., the quenched angrite Sah 99555 [1]) show excess ^{176}Hf [1-3], resulting in Lu-Hf dates significantly older than the Solar System. One possible explanation for this is that irradiation in the early Solar System produced the short-lived isomer ^{176m}Lu ($t_{1/2}$ = 3.7 hr) and thus accelerated the ^{176}Lu-decay [4-5].

In contrast to the results of Sanborn *et al.* [6], we found excess ^{176}Hf in the quenched angrite D'Orbigny and the plutonic angrite NWA 4801. Our internal Lu-Hf isochrons yield dates of 4760 ±64 Ma for D'Orbigny (8 fractions) and 4635 ±19 Ma for NWA 4801 (10 fractions). The initial ^{176}Hf/^{177}Hf values are 0.279736 (37) and 0.279788 (89), respectively, which are in the same range as the CHUR values of [7-8] back-calculated to 4567 Ma, and 3-6 ε-units higher than the initial Hf isotope composition of the Solar System suggested by Bizzarro *et al.* [1].

Our new data do not preclude irradiation on the angrite parent body as a possible cause for the old apparent ages. However, not all Lu-Hf dates are too old: our 8-point Lu-Hf isochron for the plutonic angrite NWA 4590 yields 4580 ±46 Ma, which is in agreement with the Pb-Pb age reported by [9-10]. Its initial ^{176}Hf/^{177}Hf value of 0.279794 (16) agrees with the CHUR values of [7-8] back-calculated to 4567 Ma.

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