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Activity disequilibrium of ^{230}Th , ^{234}U , and ^{238}U in old stilbite: Effects of young U mobility and daughter implantation by α -recoil

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Palaeoproterozoic stilbite from veins in the Malmberget and Svappavara (northern Sweden) ore deposits is characterized by Pb_{rad} excess relative to U, and by activity ratios of $[\text{}^{234}\text{U}]/[\text{}^{238}\text{U}] > 1$ and $[\text{}^{230}\text{Th}]/[\text{}^{238}\text{U}] > 1$. For each specimen, the data define a regression line that is steeper than the equiline in the $[\text{}^{230}\text{Th}]/[\text{}^{232}\text{Th}]$ vs. $[\text{}^{238}\text{U}]/[\text{}^{232}\text{Th}]$ diagram. The activity disequilibrium requires a disturbance of the U-Th systematics within the last 1 M.y. Leaching and infiltration experiments on Malmberget stilbite demonstrate (1) preferential leaching in the order $\text{Pb} > \text{U} > \text{Th}$ and uptake in the order $\text{Th} > \text{Pb} > \text{U}$; and (2) isotopic fractionation of U by preferential mobilization of ^{238}U and ^{235}U relative to ^{234}U . Stepwise-leaching experiments indicate that the bulk of U is hosted in the channel sites of stilbite. The Th-U disequilibrium systematics can be explained by (1) addition of U with $[\text{}^{234}\text{U}]/[\text{}^{238}\text{U}] > 1$ from a fluid or alternatively (2) loss of U from a two-component system that consists of an open and a closed component. U addition requires a multi-stage history for all analyzed stilbite samples that involved multiple gain or loss of U and/or Pb. In contrast, U loss does not require any multi-stage process but can be explained by preferential removal of ^{238}U and ^{235}U relative to recoiled daughter isotopes such as ^{234}U , ^{230}Th , and Pb_{rad} during a single event. This requirement implies an open-system behavior for the channel sites and a closed-system behavior to the silicate framework of stilbite. Each α -recoil directly or indirectly, i.e., through its recoil cascade, damages the silicate framework. Subsequent annealing of the damaged stilbite lattice may result in immobilization of the recoiled daughter isotopes. In this model, the combination of U loss with reduced mobility of recoil-displaced daughter isotopes accounts for the observed Th-U disequilibrium systematics, the similarity in Th/U as deduced from Th-U disequilibrium and Pb isotope data, and the Pb_{rad} excess. The closed-system could in part be represented by such immobile recoiled material. These two contrasting explanations involve either multi-stage or multi-component systems. None of the models permits the derivation of an accurate age.

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Uranium (VI) uptake by calcium carbonate and lake sediments

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Uranium migration in natural aqueous systems is an ongoing concern in environmental research. Sorption interactions with soils, sediments and rocks are important mechanisms for understanding the uranium mobility and correction of U/Th dating methods for open systems. Uranium immobilization is possible due to reduction U (VI) to U (IV), adsorption or co-precipitation.

Under oxidizing environmental conditions, uranium typically occurs in the hexavalent form as the mobile, aqueous uranyl ion (UO_2^{2+}) [2]. Moreover, depending from environmental conditions, uranium forms carbonate complex such as $\text{UO}_2(\text{CO}_3)_2^{2-}$ or $\text{UO}_2(\text{CO}_3)_3^{4-}$ [1].

Uptake of such dissolved metal contaminants by many fine-grained mineral phases (clays, oxides, and hydroxides) is most commonly achieved by adsorption. For carbonates recent evidence suggests that incorporation into the solid (co-precipitation) dominant uptake.

In this study we have conducted laboratory experiments of uranium (VI) sorption by lake sediments and by calcium carbonate. The sediment samples were obtained from an artificial lake (South-West Germany). Experiments were conducted under O_2 and CO_2 free atmosphere and in pH range 2-11. Results from the study suggest that uranium uptake by this sediment and calcium carbonate is strong depending from pH, and that low and high pH is minimal, and the maximum amount of Uranium sorption occurs near neutral pH. For calcium carbonate uranium uptake at pH 7 is close to 100 % and depends from solution compound and surface area.

References

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