

On the fate of ^{220}Rn in partially saturated media

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Thoron (^{220}Rn , half-life 55.6s) is a short lived isotope of the radioactive noble gas radon (main isotope: ^{222}Rn , half-life 3.58d). While ^{222}Rn is part of the ^{238}U -decay series, ^{220}Rn originates from the ^{232}Th -decay series. As 220 , ^{222}Rn are produced from alpha-decay of radium isotopes, 220 , ^{222}Rn emanate from minerals containing their precursors 224 , ^{226}Ra . As a result, both radon isotopes are found in soil gas and soil-near air, ^{222}Rn also in ground water. Hence, it is expected to find also ^{220}Rn in ground water. However, ^{220}Rn could not be detected in natural aquatic environments, although ^{220}Rn in water can be measured in laboratory experiments by a tailored improved radon- and thoron-in-water detection system.

In a field experiment, 220 , ^{222}Rn concentrations in various depths were continuously monitored in the unsaturated soil. During a rain event, the lower most sampling port was flooded by the rising groundwater table. In response to saturation, the ^{222}Rn concentrations decreased by 40-50%, but the ^{220}Rn concentrations become virtually zero. After the event, e.g. the return to partly unsaturated conditions, the ^{222}Rn concentrations increased to former levels whereby the ^{220}Rn concentrations only slightly rose to 20% of the pre-event concentration. We conclude from the different response of 220 , ^{222}Rn , that the release of ^{220}Rn to soil air is controlled by the soil water content, i.e. the thickness of water films around the soil grains. As the diffusion length of ^{220}Rn is about 75 times smaller than that of ^{222}Rn , most of the short lived ^{220}Rn decays within the water film, if the layer reaches a certain critical thickness. In contrast, the long lived ^{222}Rn diffuses through the water film into the soil air.

We tested this hypothesis in laboratory experiments with Mn-coated sand, that emanates ^{220}Rn . In a closed vessel, the sand was sequentially wetted while the ^{220}Rn concentration in the surrounding air was analyzed. Preliminary results show that the release of ^{220}Rn from sand to air is effectively controlled by the water content of the sand: at a critical water content, the measured ^{220}Rn concentration dropped to almost zero, adding experimental support that the release of ^{220}Rn is controlled by the available humidity in the porous media.

A new highly effective silicagel emitter for lead isotopic measurements

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In modern U-Pb-geochronology a precision as high as 0.01% can be achieved in isotopic analyses using thermal ionization mass spectrometry. One of the prerequisites for achieving this high precision was the development of a highly effective ion emitter [1], which is based on a colloidal silicagel formerly produced by the chemical company Merck. Unfortunately this silicagel is not available for purchase anymore, and another ion emitter will be needed if the emitter efficiency of colloidal silica deteriorates with time. We tested colloidal silicagels from Alpha Aesar, Nissan Chemicals and Sigma-Aldrich as possible alternatives to the established Merck emitter. The gels were tested for U and Pb blanks and the ion yield (number of ions detected divided by number of atoms loaded) depending on the SiO_2 concentration and particle size using 300 pg Pb of the Pb isotopic standard SRM-981. For the most effective emitter, we also searched for the optimal mixing proportion of silicagel and H_3PO_4 .

The Pb blanks of the gels from all three companies are below 0.7 pg/g and for Sigma-Aldrich ~0.15 pg/g. The U blanks from Nissan Chemicals are between 21 and 38 pg/g and for Alfa Aesar ~0.7 pg/g, rendering these silicagels unsuitable for U analysis. The gel from Sigma-Aldrich shows low U blanks below 0.1 pg/g. We found no correlation between ionisation efficiency and the particle size of SiO_2 . The most effective emitter is from Sigma-Aldrich at a concentration of 0.4%. The average ion yield for this concentration is 5.8% (n=18), higher than the average ion yield of 4.1% (n=23) of the Merck gel. The accuracy and reproducibility of the isotopic ratios of both gels are comparable ($^{207}\text{Pb}/^{206}\text{Pb} = 0.914740 \pm 0.000041$ for Sigma-Aldrich and $^{207}\text{Pb}/^{206}\text{Pb} = 0.914735 \pm 0.000048$ for Merck). Fractionation factors range between 1.0005 and 1.0012. The highest ion yield for Sigma-Aldrich gel was obtained using 92 μg of H_3PO_4 and 0.004 ml of gel.

From these observations we conclude that the Sigma-Aldrich silicagel is an excellent alternative for the established Merck silicagel for Pb isotopic measurements.

[1] Gerstenberger & Haase (1997), *Chem Geol* **136**, 309-312.