Source and speciation of ^{14}C in a cement-based repository for radioactive waste

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Carbon-14 is an important contributor to the annual dose released from a cement-based repository for low- and intermediate-level radioactive waste in Switzerland. To date, performance assessment has to be based on assumptions regarding the speciation and mobility of ^{14}C bearing compounds. It is considered that ^{14}C mainly contributes to dose in its organic form, such as ^{14}C bearing organic compounds, which are only weakly retarded in the cementitious near field.

Compilations of the activity inventories reveal that, in the already existing and future arising of radioactive waste in Switzerland, the ^{14}C inventory is mainly associated with activated steel (~ 85 %). The inventories of activated steel and ^{14}C in the repository are well known. However, the chemical speciation of ^{14}C in the cementitious near field upon release from activated steel is only poorly investigated.

Identification and quantification of the ^{14}C bearing organic compounds formed during the anoxic corrosion of activated steel is a major challenge, firstly due to the low ^{14}C inventory of the material available for the corrosion experiment, and secondly due to the extremely low corrosion rate of steel in alkaline solution (few nm/year). Thus, the development of an analytical method with a very low ^{14}C detection limit is required, such as compound-specific ^{14}C AMS. As a first step towards the development of the technique, batch-type corrosion experiments with non-activated, carbon-containing iron powders were carried out with the aim of identifying potentially ^{14}C bearing organic compounds. The powders were immersed in groundwater- and cement-type solutions and time-dependent changes in the concentrations of dissolved and gaseous carbon species were determined using high-performance ion exclusion chromatography coupled to conductivity detection and mass spectrometry and gas chromatography coupled to mass spectrometry. The conceptual approach for a corrosion study with activated steel along with first results will be discussed.

Irradiation histories of meteoritic inclusions measured by ^{40}K

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The solar and galactic cosmic rays recorded by chondrules and refractory inclusions (CAIs and AOAs) can provide unique information on their formation, transport and storage histories. To explore the irradiation histories of these objects, we have developed novel analytical protocols to accurately measure Ca/K ratios and K-isotopes by thermal ionization mass spectrometry [1]. The low abundance of ^{40}K relative to ^{39}K makes the $^{40}K/^{39}K$ ratio a sensitive indicator of spallation, whereby efficient spallogenic production of ^{40}K occurs through proton irradiation of calcium targets by $^{43}Ca(p,\alpha)^{40}K$ and $^{44}Ca(p,n+\alpha)^{40}K$. Because proton stopping and reaction depths at relevant energies are similar to the sizes of the inclusions, ^{40}K depth profiles can allow us to discriminate between solar and galactic cosmic rays. Further, the common K retains information on precursor irradiation.

We selected well characterized CAIs and AOAs from Efremovka carbonaceous chondrite (CV3red) and chondrules from the NWA 5697 ordinary chondrite (LL3). The three chondrules selected for study have variable U-corrected Pb-Pb dates and ^{54}Cr compositions [2], suggesting lateral transport and storage of these objects in the solar protoplanetary disk for at least ~2 Myr prior to their accretion in the NWA 5697 parent body. All material analyzed show large (i.e. permil to percent) positive ^{40}K anomalies that broadly correlate with Ca/K ratios, consistent with spallation of Ca. The ^{40}K anomalies in the 3 chondrules vary with their absolute Pb-Pb dates, such that the oldest chondrules register the largest doses. At face value, this is consistent with a monotonic irradiation source, suggesting that ^{40}K anomalies were generated during storage in the protoplanetary disk, likely from exposures to galactic cosmic rays. The comparable dosages recorded by CAIs and chondrules suggest a similar transport mechanism, possibly associated with stellar outflows [3]. Similarly to CAIs, we infer that some chondrules formed in the inner regions of the protoplanetary disk were ejected to large orbital distances and drifted back to the accretion regions of chondrites over timescales of a few Myr. We are currently conducting ^{40}K depth profiles of large, well characterised and Pb-Pb dated chondrules to test this conclusion.

[1] Wielandt & Bizzarro (2011) *JAAS* **26**, 366 [2] Connelly *et al.* (2012) *Science* **338**, 651 [3] Shu *et al.* (1996) *Science* **271**, 1545

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