Extractions of *in-situ* produced ^{14}C in terrestrial quartz and measurements of ^{14}C production rate using saturated quartz

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The measurement of cosmic-ray-produced [10]Be and [26]Al in quartz exposed at the Earth's surface has enabled numerous scientific investigations aimed at quantifying landscape evolution. We are establishing a reliable measurement system for *in-situ* produced [14]C ($t_{1/2}$=5,730 yr) extraction from quartz, based on step heating of powdered quartz samples to 500-1550°C under flowing carrier gas in a tube furnace. The background of the current extraction system is $(2-5) \times 10^5$ atoms ^{14}C using deeply shielded quartz from Homestake mine. This is comparable to the background level reported by Lifton *et al.*, (2001) using their "Dry extraction" method. The recovery yield of carrier gas is >95%.

To obtain the production rate of *in-situ* produced ^{14}C in quartz, we measured ^{14}C in purified quartz from the Beacon sandstone collected from the Transantarctic Mountains, Antarctica. Since [10]Be and [26]Al concentrations in aliquot of samples were nearly saturated, the erosion rates of these samples were less than a few mm/10[4] yr. *In-situ* produced ^{14}C in the quartz, therefore, is saturated impling that observed ^{14}C activity in the quartz sample is equal to the production rate of ^{14}C in the quartz for the ^{14}C mean-life. This is the most direct and unambiguous method to obtain the production rate of a radionuclide.

The preliminary result is $(8.2\pm0.5) \times 10^5$ atom ^{14}C/g SiO_2 compared to $(6.4\pm0.1) \times 10^7$ atom [10]Be and $(2.2\pm0.1) \times 10^8$ atom [26]Al/g SiO_2 respectively. The preliminary ^{14}C/[10]Be and ^{14}C/[26]Al production rate ratios at the high elevation site are 2.4 and 0.4, respectively.

Comparison of our preliminary results for ^{14}C in saturated Antarctic rocks and the most recent estimate of the production rate from Lifton *et al* (2001) indicates that our *in-situ* produced ^{14}C extraction efficiency from quartz is greater than 94%.

Reference

Lifton N.A., Jull A.J.T. and Quade J., (2001). *Geochim. Cosmochim. Acta* **65**. 1953-1969.

A redox front migration process in sedimentary rock- Long-term behavior of nuclide migration relevant to near-field processes in radioactive waste disposal -

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The development of redox fronts in geological environments, e.g. in the near- and far-fields of high level radioactive waste (HLW) repositories of all designs, will be unavoidable. The long-term behaviour of redox front migration, therefore, is one of the key processes to evaluate the barrier function of the near-field geological environment.

As an analogous example of long-term redox front migration, investigations have been carried out on a redox front generated up to several meters below the ground surface in Tertiary sedimentary rock occurring in central Japan. The geological setting and features of the redox front's spatial distribution pattern show that redox reactions have progressed within approximately the last several hundreds of ka, following development of the present surface topography of the Quaternary cover. Detailed microscopic observations and geochemical analysis by XRF, EPMA, ICP-MS and SEM-EDS show that the redox front has accumulated concentrations of major and minor elements that are at least several times more than the background levels. Particularly, transition elements such as Zn, Co and Ni as well as HREE are co-precipitated within a band where Fe and Mn oxide are concentrated at the front. Microscopically, aggregations of spheroidal amorphous ferric iron grains with diameters of a few micrometers are identified in the pore spaces of the sedimentary rock. This suggests that the migration of the redox front might occur at the same time as these amorphous iron oxide grains move through the rock matrix scavenging the dissolved elements during their migration, probably due to their high effective surface area which is available for ionic adsorption. Such a process could also be effective for radionuclide retardation in the near field environment. The profile of ratios of ferrous to ferric iron, from the oxidising part to the reducing part through the front, also reveals that the oxidised part of the host rock has slightly recovered its reducing environment after the redox front has passed. This is suggested by the supply of reducing pore water through the surface organic rich soil, and the mobile morphological feature of oxidants might also be considered as a reason for redox front movement.

The front is considered a long-term analogue for a geochemical process expected to occur in the sedimentary rock that might occupy the near-field environment of a future HLW repository.