



Contribution ID: 161

Type: Verbal

Speciation of Carbon-14 in a Cementitious Repository for Radioactive Waste: Identification of Organic Compounds in Anoxic Corrosion Experiments with Iron Powders

Thursday, 15 May 2014 09:30 (15 minutes)

Carbon-14 is an important contributor to the annual dose released from a cement-based repository for low- and intermediate-level radioactive waste (L/ILW) in Switzerland. In current performance assessment it is assumed 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 in L/ILW is mainly associated with activated steel (~85 %). In nuclear power plants ^{14}C is produced by the activation of nitrogen impurities in stainless steel exposed to thermal neutron fluxes (e.g. in nuclear reactors) according to the reaction $^{14}\text{N}(n,p)^{14}\text{C}$. The chemical speciation of ^{14}C in the cementitious near field upon release from activated steel is only poorly known.

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 activated steel, and secondly due to the extremely low corrosion rate of stainless steel in alkaline solution (few nm/year). Thus, conducting a corrosion study with activated steel requires the development of an analytical method with a very low ^{14}C detection limit for the identification and quantification of the ^{14}C bearing organic compounds released during the anoxic steel corrosion, such as compound-specific ^{14}C accelerator mass spectrometry (AMS).

As a first step towards the development of the AMS-based analytical 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 iron powders were immersed in artificial cement pore water solutions in zero-headspace, gas-tight vials and time-dependent changes in the concentrations of low molecular weight (LMW) dissolved and volatile carbon species were monitored over a period of 35 days. Dissolved and volatile organic compounds were determined in the supernatant solution using high-performance ion exclusion chromatography (HPIEC) coupled to conductivity detection (CD) and mass spectrometry (MS) for the dissolved species and headspace gas chromatography (GC) coupled to mass spectrometry for the volatile compounds. Organic carbon species were identified to be volatile hydrocarbons, like methane, alcohols, aldehydes, and carboxylic acids, with a maximum of up to four carbon atoms. Results from the corrosion experiments with non-activated iron powders will be discussed in detail along with the conceptual approach for the future corrosion study with activated steel.

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Session Classification: Chemistry of Nuclear Fuel Cycle 3

Track Classification: Chemistry of Nuclear Fuel Cycle / 1st ASGARD International Workshop