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Retention of ^{14}C small organic molecules in cementitious environments

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Due to its half-life ($t_{1/2}$ = 5730 years) and expected potential mobility, ^{14}C is a safety-relevant radionuclide in the context of repositories for low- and intermediate-level radioactive waste (L/ILW). The largest contribution to the ^{14}C inventory is produced during reactor operation by the activation of ^{14}N ($^{14}N(n,p)^{14}C$) with thermal neutrons. ^{14}C can be released both as organic and inorganic components, depending on the nature of the waste material and the chemical conditions of the near field. Hence, low-molecular-weight ($C \leq 5$) organic molecules containing ^{14}C (e.g., alkanes, alcohols, aldehydes, or carboxylic acids) can be released as a result of the corrosion of steel components. In spite of the key role of cement in repositories for L/ILW, only a limited number of experimental studies dealing with the retention of low molecular-weight organics with cementitious materials is available to date in the literature, with partly contradictory results.

This work aims at investigating the sorption behavior of representative low-molecular-weight organics by selected cementitious materials and cement phases. Experiments are performed under Ar-atmosphere using a combination of ^{14}C-labelled and inactive compounds, i.e., n-pentane, n-propanol, acetaldehyde, acetone, formic and citric acid. The cementitious materials investigated include CEM I (Portland cement), CEM III/B (with ≥ 66% slag content) and calcium silicate hydrate (C-S-H) phases with Ca : Si ratio of 0.8 and 1.5. The hardened cement pastes resulting from CEM I and CEM III/B are characterized by significantly different content of C-S-H and AFm phases, which are expected to play a key role in the retention of the investigated organic systems. Solid-to-liquid ratios are varied within 10-100 $g \cdot L^{-1}$ due to the expected weak retention of some of the investigated organic compounds. Quantification of the organics retention is achieved both by liquid scintillation counting (LSC, ^{14}C-labelled samples) and dissolved organic C content (NPOC, inactive samples). The main outcome of this comprehensive sorption study will be presented in this contribution.

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