



# Sorption and diffusion studies with low molecular weight organic compounds in cementitious systems



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## ARTICLE INFO

### Article history:

Received 14 October 2015

Received in revised form

25 January 2016

Accepted 30 January 2016

Available online 2 February 2016

### Keywords:

Cement

Diffusion

Inverse modelling

Sorption

Alcohols

Aldehydes

Carboxylic acids

Carbon-14

## ABSTRACT

The uptake of methanol, ethanol, formaldehyde, acetaldehyde, formic acid and acetic acid by cement paste was determined in hydrating cement after 1 h, 28 d and 390 d hydration. The sorption values determined for formate and acetate were critically assessed by investigating through- and out-diffusion of these compounds in fully hydrated cement paste and their uptake by individual cement phases. Diffusion studies included inverse modelling of four data sets for each compound and an uncertainty analysis based on a Latin hypercube sampling procedure. Solid–liquid distribution ratios determined from the hydration experiments are on the order of  $10^{-4} \text{ m}^3 \text{ kg}^{-1}$  in the case of alcohols and aldehydes indicating non-specific (very weak) bonding onto the surface of the cement phases, e.g. through hydrogen bonding. Hydration and diffusion studies reveal slightly higher distribution ratios and reversible uptake by cement paste and cement phases in the case of acetate indicating specific adsorption (electrostatic interaction) onto partially positively charged surface sites of the cement phases. Selective binding of a small fraction of formate is evidenced from both sorption and out-diffusion experiments suggesting the presence of sorption sites capable of strongly bonding the anion, presumably by  $\text{SO}_4^{2-}/\text{HCOO}^-$  replacement in the ettringite structure.

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## 1. Introduction

Carbon-14 is a radionuclide of particular interest in nuclear power production and a key radionuclide in performance assessment (PA) of deep geological repositories for radioactive waste (e.g. Nagra, 2002; Yim and Caron, 2006; Johnson and Schwyn, 2008; Nuclear Decommissioning Authority, 2012). The half-life of  $^{14}\text{C}$  (a beta emitter) is sufficiently long (5730 y) for its release to be of relevance after repository closure and its behaviour in the repository system is of specific concern for several reasons: i) The release of  $^{14}\text{C}$  from the disposal facility may occur as dissolved or gaseous species, ii) The mobility of the  $^{14}\text{C}$  containing organic compounds is expected to be very high due to weak interaction with the minerals of the host rock under near-neutral conditions, and iii)  $^{14}\text{C}$  can be taken up by biota after mixing into the cycle of stable  $^{12}\text{C}$  and  $^{13}\text{C}$ . The importance of  $^{14}\text{C}$  in the safety case further arises from specific assumptions that have to be made in connection with PA due to the currently very limited information available on the  $^{14}\text{C}$  speciation, the fate and retention of  $^{14}\text{C}$  in the multi-barrier system and the rate of release into the host rock

(Johnson and Schwyn, 2008). For example, it is presently assumed that  $^{14}\text{C}$  containing organic compounds are only weakly retarded by the cementitious near field of a repository for low- and intermediate level waste (L/ILW) or, conservatively, even no retardation has been considered (Nagra, 2002).

The main sources of  $^{14}\text{C}$  in radioactive waste from light water reactors (LWR) in operation in Switzerland are activated metallic nuclear fuel components, reactor core structures components, spent filters and ion exchange resins used in the LWRs for the removal of radioactive contaminants in reactor coolant. In LWRs,  $^{14}\text{C}$  is the product of mainly nitrogen-14 activation ( $^{14}\text{N}(n,p)^{14}\text{C}$ ), which occurs by exposing  $\text{N}_2$  containing metals to thermal neutron flux (Yim and Caron, 2006). Compilations of the activity inventories reveal that, in the already existing and future arising of radioactive waste in Switzerland, the  $^{14}\text{C}$  inventory is mainly associated with activated steel while the contribution from spent ion exchange resins and filters, Zircaloy and waste forms from medicine, industry and research to the  $^{14}\text{C}$  inventory are minor (Nagra, 2014). Note that spent fuel and high-level waste are foreseen to be disposed of in a separate repository in Switzerland.

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