Coprecipitation of 14C and Sr with carbonate precipitates: The importance of reaction kinetics and recrystallization pathways

DAVID J HODKIN[1] DOUGLAS I STEWART[2] JAMES T GRAHAM[3] IAN T BURKE[1*]

[1]School of Earth and Environment and [2] School of Civil Engineering, University of Leeds, Leeds, UK, LS2 9JT

[3]National Nuclear Laboratory, Sellafield, UK, CA20 1PG

^{90}Sr and ^{14}C are two of the contaminants found at elevated levels in groundwater under the Separation Area of the Sellafield reprocessing site in Cumbria, UK. A conventional remediation technique for this suite of contaminants may involve pumping the groundwater and passing it through anion and cation exchange media. However, the effectiveness of this process is reduced by competing ions present in groundwaters (e.g. Ca^{2+}, Na^{+})[1].

Precipitation of ^{90}Sr and ^{14}C as insoluble carbonate[2] offers a potentially lower cost alternative, producing a solid residue that is readily grouted in cement wasteforms.

It has been shown that the crystallization pathways occurring between aqueous ions of Ca^{2+} and CO_3^{2-}, and thermodynamically stable calcite are an important control on the removal of ^{14}C from solution. If the precipitate undergoes recrystallization ^{14}C and ^{90}Sr will become remobilized to the solution. This is of minimal importance for the Sr, which is reprecipitated into the newly formed crystal lattice. However, in systems open to atmosphere, ^{14}C undergoes mixing with ^{12}C derived from in-gassing of CO_2 and becomes diluted, reducing its removal efficiency.

Solution $Ca:CO_3$ ratios were found to be important for ^{14}C removal. Where CO_3^{2-} was present equal to or in excess of Ca, ^{14}C removal was limited due to the dilution of $^{14}CO_3^{2-}$ by $^{12}CO_3^{2-}$. Sr removal however was not significantly affected, as atmospheric CO_2 in-gassing was able to provide sufficient CO_3^{2-} for full Ca^{2+}, and thus Sr^{2+} removal. This process was able to achieve maximum removals of 99.7% for ^{14}C and 98.6% for ^{90}Sr in the $Ca[10mM]:CO_3^{2-}[1mM]$ experiment.

Extrapolating these removal efficincies from our experiments to the most contaminated groundwater present at Sellafield suggests that each m^3 of groundwater would produce 1.23 kg of calcite containing 123.23 MBq of β activity (75 GBq/ton). The effluent from this process would contain a residual 0.15 KBq L^{-1} of ^{14}C and 0.62 KBq L^{-1} of ^{90}Sr.

[1] Marinin D & Brown G. 2000 Waste Manag. 20 (7)
[2] Fujita et al. 2000 Geomicrobiol. J. 17 (4)