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Retention of $^{14}CO_3^{2-}$ by hydrated cement and calcite: impact of ISA and chloride

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^{14}C is produced from stable parent isotopes (^{13}C, ^{14}N, ^{17}O) present in different components of nuclear reactors. Steel components containing ^{14}C are disposed in repositories for low and intermediate level wastes (L/ILW). ^{14}C can potentially be released as small organic molecules or $^{14}CO_3^{2-}$. Retention mechanisms proposed for the uptake of $^{14}CO_3^{2-}$ in cement-based repositories for L/ILW include adsorption onto cationic surfaces of C-S-H phases or isotopic exchange with non-radioactive carbonate phases present in cement [1]. Calcite itself is considered as the final degradation stage of cement, occurring once C-S-H phases are dissolved. This work investigates the effect of ISA and chloride on the uptake of $^{14}CO_3^{2-}$.

Batch sorption experiments with hydrated cement CEM I at pH ≈ 13.6 were conducted under Ar atmosphere. Sorption experiments with calcite were performed under air at pH ≈ 8.3. Experiments were conducted by systematic variation of (i) solid-to-liquid ratios (1–50 $g \cdot L^{-1}$), (ii) initial concentrations of $^{14}CO_3^{2-}$ ($8.9 \cdot 10^{-11}$–$8.6 \cdot 10^{-9}$ M), (iii) equilibration time (2–140 days), (iv) ISA concentrations ($1 \cdot 10^{-5}$–0.2 M), and (v) chloride concentrations ($1.0 \cdot 10^{-4}$–2.0 M). Phase separation was achieved by ultrafiltration or ultracentrifugation. The concentration of ^{14}C was quantified by liquid scintillation counting.

The uptake of $^{14}CO_3^{2-}$ by cement is fast and moderately strong, with $R_d \approx 10^3$ $L \cdot kg^{-1}$. Sorption experiments reflect a strong dependency of the uptake on the initial concentration of $^{14}CO_3^{2-}$, which is attributed to isotopic exchange processes. Chloride has a negligible impact on the uptake of $^{14}CO_3^{2-}$. A decrease in R_d related to ISA is observed only at $[ISA] > 10^{-2}$ M. In the case of calcite, the overall uptake strongly depends on the equilibration time. At short equilibration times, $^{14}CO_3^{2-}$ is mainly adsorbed on the surface of calcite particles. A systematic increase of the uptake is observed with time, caused by the recrystallization of calcite and isotopic exchange processes. Chloride and ISA have a negligible impact on the retention of $^{14}CO_3^{2-}$ under the investigated boundary conditions.

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References

1. M. Ochs, D. Mallants, L. Wang (2016), Springer, 301pp.