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Anaerobic biodegradation of citric acid in the presence of cement pellets: Impact on cement integrity

NATALIE BYRD[1], KATHERINE MORRIS[2], **ANANYA SINGH**[1], JOE SMALL[2], FRANK TAYLOR[3], CHRISTOPHER BOOTHMAN[1], DIRK ENGELBERG[2], TRISTAN LOWE[1], SULTAN MAHMOOD[1] AND JONATHAN RICHARD LLOYD[1]

[1]The University of Manchester

[2]University of Manchester

[3]Nuclear waste services

Presenting Author: ananya.singh@manchester.ac.uk

Understanding potential physical and biogeochemical mechanisms that impact on Low Level Radioactive Waste (LLW) disposal systems is essential for disposal saftey case design. Cement cracking may impact contaminant migration and a key mechanism for crack self-sealing is CaCO3 precipitation. In LLW this could be facilitated by microbial oxidation of organics, including citrate, which produces CO32-/HCO3- species. We investigated the biodegradation of citrate, a widely used decontamination agent, in the presence of cement pellets to explore impacts of biodegradation on cement evolution. Nitrate-reducing microcosms containing cement were initiated at pH 11.2 and periodic samples underwent geochemical analyses by pH, ion chromatography and ICP-AES. Citrate-supplemented microcosms showed complete citrate removal in 40 days, accompanied by denitrification, acidification and a decrease in Ca2+(aq) concentration. By day 40, visible precipitates had formed on cement pellets in citrate-supplemented microcosms; these pellets were analysed by SEM, EDS and 3D XCT, and XRD. Data from SEM, EDS and XRD indicated the precipitates coating the cement were calcite. The 3D XCT analysis of the cement samples from citrate-supplemented microcosms showed calcite had sealed surface cracks . In the no electron donor control, there were no detectable changes in aqueous geochemistry, the surface mineralogy appeared unaltered, and cracks were not sealed by day 40. Findings here suggest citrate was oxidized to HCO3-, which reacted with Ca2+(aq) from cement to form calcite precipitates on the cement surface, resulting in crack-sealing. This is likely a positive prospect in terms of reducing contaminant mobility: (1) by removing citrate, a potential complexant in LLW and (2) by producing CO2, which forms COCO3- species that can react with Ca2+(aq) from cement to promote cement crack-sealing.