The effect of microbial activity on the near and far fields of a Swiss low/intermediate level waste repository

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Introduction

A three year experimental programme was undertaken for Nagra, Switzerland to investigate the gross effects of microbial activity on repository geochemistry, radionuclide sorption and the integrity of repository and host rock materials. This work was carried out to quantify the effects of microbial activity on the deep disposal of low and intermediate level waste arising from a Swiss 120 GWa nuclear energy scenario [1].

Methods

The investigation was facilitated by a series of batch laboratory experiments the aims of which were to simulate the near-field waste environment (over a period of time) by using a system of eight batch-reaction cells containing waste, containment and backfill material inoculated, in some cases, with microbiological cultures in order to ascertain the influence of microbes on the near-field. The cells were monitored for changes in pH, Eh, leachate and gas production, colloid composition and numbers and microbial numbers with time. Changes in physical integrity of the components were assessed at the end of the experiment. In addition, the effect of microbial presence on sorption processes in the far-field was assessed. Details of the cell construction and all methodologies are given elsewhere [2]. Four types of material were supplied by Nagra for use in the experiments: carbon steel coupons representing waste canister material; ion-exchange resins (Powdex Inactive) embedded in Portland cement paste representing waste packing; Portland cement-pumice composite blocks representing backfill material; and marl core from the Oberbauenstock Marl representing host rock lithology. The cells were filled with 2 l of a synthetic groundwater designed to simulate the conditions likely to be encountered in a potential Swiss repository.

Results

The cells were run for ~550 days after which five of the cells were opened for examination. The remaining three cells were opened after ~1730 days. Analytical results over 550 days showed that the pH in all cells became alkaline averaging 11.5–12 with the pH stable. Eh (mV vs SHE) fluctuated in all cells but monitoring indicated a general decline to an average –100mV. A large dataset was generated from leachate analysis and is detailed elsewhere [3] but from inspection some trends are clear. The chemistry of the leachate generated in these cells appears to be dominated by cement hydration and degradation reactions giving a high pH with varying concentrations of Ca, Mg, total alkalinity, Ba, Al and Fe, and increasing concentrations of Si, B, K, Total Organic Carbon (TOC), and F with time. Microbiological results show fluctuations in microbial populations in all the cells with time. Thus an alkalotolerant population has developed in the low nutrient conditions present. Many of the observed changes in the fluid chemistry can be correlated to reactions exhibited by the solid components and to the precipitation of secondary phases. In particular, early changes (0–100 days) in fluid chemistry can be explained by the precipitation of talc and calcite which resulted in lower buffering of pH (*c.* 10–10.5), removal of Mg and lowering of Ca in the fluid. After exhaustion of Mg in the fluid, the pH was buffered by cement hydration reactions (of calcium silicate hydrate, CSH, gels) and rose to 11.5–12. Steel coupons all exhibited signs of corrosion. Estimates of the rate of corrosion by metal weight loss indicated that the overall corrosion rates are at the low end of the range expected for aerobic corrosion. However, detailed petrographic observations revealed that corrosion was localised in deep crevice-pits. Some coupons had already been penetrated to 50% of their thickness by these localised corrosion features. This suggests that the coupons could be breached two orders of magnitude faster than indicated by the average corrosion rate determined by metal weight loss measurements.

The remaining three cells show the development of microenvironments where the interactions of aerobic and anaerobic biogeochemical processes have led to further evolution of the systems.

Conclusions

This multidisciplinary study has generated a unique and large database. A methodology has been established for examining gross changes in a repository system and generating useful data relatively quickly. Although the study was aimed at examining microbial effects on a Swiss intermediate/low level waste repository, the findings have a wider significance. After 550 days, the presence of microbes in these experiments did not appear to influence the overall geochemistry in any of the opened cells, although localised effects were indicated. The mineralogical studies have indicated a number of areas of concern. Steel corrosion data from normal metal weight loss methods gave a misleading impression of the behaviour of steel in the experiments. Petrographic analysis showed that corrosion is localised in deep crevice pits and that the coupons could be breached twice as quickly than indicated by weight loss measurements. This may indicate a microbial influence. The precipitation of talc and Mg-rich clay minerals in the cells could have serious implications for the long term stability of cements by lowering ambient pH through removal of OH^- causing dissolution of CSH compounds. The final 3 cells are now being characterised but already show further evolution of biogeochemistry of the cells.

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References

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