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THE IMPACT OF A (HYPER)ALKALINE PLUME ON (FRACTURED) CRYSTALLINE ROCK

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Introduction

As hyperalkaline cement porewaters leach out of the near-field, they may interact significantly with the repository host rock (e.g. Haworth et al., 1987). In consequence, those features for which the formation was originally chosen, such as low groundwater flux and high radionuclide retardation, may deteriorate. Here, available data of relevance to the interaction of repository hyperalkaline plumes on crystalline host rocks are briefly reviewed and areas of future effort identified. As most crystalline rocks may be expected to be fractured to a greater or lesser degree, focus is on fractured systems.

Evidence

Laboratory experiments

Among the earliest experiments conducted on crystalline rock were those of Bateman et al. (1999), where a range of model materials and rock types (including Avrö granite and Borrowdale Volcanic Group 'fault rock') were examined. Reaction of the original rock resulted in the development of one or more of the CSH, CASH, CKSH and CK(A)SH secondary phases which resulted in a decrease in overall porosity and an increase in surface area. Note, however, that the rock samples were crushed and the system heated to 70°C.

Of perhaps more direct relevance, a hyperalkaline fluid core infiltration experiment was performed with a sample from a fracture in Nagra's Grimsel Test Site (GTS) where the fault gouge had been preserved *in situ* (Mäder et al., 2006). Over nine months, a gradual decrease in flow rate by a factor of 25 was observed and was attributed to clogging of flow paths in the fracture by secondary CSH precipitates due to reaction of the rock with the hyperalkaline fluids.

Field (URL) experiments

What little work that has been carried out in URLs to date has not yet been published comprehensively enough to allow full integration within this short note. The most ambitious (and successful) experiment to date has been the international HPF (Hyperalkaline Plume in Fractured Rock) project at the GTS. This long-term field experiment was carried out by injecting hyperalkaline fluids into a water-conducting shear zone along with parallel, *in situ* radionuclide transport experiments and supporting laboratory experiments (see above). Although only a short overview (Mäder et al., 2006) has been published to date, the full report is expected soon. Briefly, the results indicate that:

- hyperalkaline fluids cause significant dissolution of the primary mineralogy of the host rock and precipitation of secondary minerals that induce changes (including self-sealing) to the flow field
- the conceptual model (cf. Alexander and Smellie, 1998) for the likely evolution of a hyperalkaline plume in crystalline (and other) host rocks is valid

Natural analogues (NA)

The most directly relevant NA study to date was that carried out across the various sites in Jordan (see Alexander and Smellie, 1998; Pitty, 2009 and references therein for details). Although the host rock was a clay biomicrite, the data on the spatial and temporal changes to the host rock following reaction with the hyperalkaline plume (max. pH measured of 12.9) are transferable (with care) to fractured crystalline rock (and clays, see the comments in Mazurek et al., 2006). The main quantitative conclusions were that the conceptual model for alteration of the host rock was largely correct and that reaction will generally seal fractures (cf. HPF above). In addition, comparison of the alteration of the clay biomicrite and locally abundant basalt indicated that the secondary mineral assemblage was generally very similar. Currently, two new NA studies (Alexander et al., 2008; Milodowski et al., 2009) on low alkali cement leachate interaction with bentonites are ongoing and both are expected to produce information on reaction in fractures in the crystalline (ophiolite) basement beneath the bentonites.

Model development

Modelling of hyperalkaline fluid reaction in fractured rock began in the 1980s (e.g. Haworth et al., 1987) and, broadly, the present coupled calculational tools are adequate, although improvements could still be made (see comments in Soler et al., 2006). To date, flow and transport modelling of processes within the plume has only been attempted within the HPF project and the results were mixed, perhaps because of the highly complex nature of the experimental fracture.

A special case: grouts

As noted above, most crystalline rocks will be fractured to some degree so, to minimise groundwater ingress, some degree of fracture sealing, most probably by grouts, will be necessary. Concerns were raised (e.g. Vieno et al., 2003) when it was realised that several hundred tonnes of grouts could easily be required in a typical crystalline rock repository and that the hyperalkaline leachates from these grouts could impact the repository. This, in part, prompted the re-examination of previous work on low alkali cements (due to the generally lower pH of any subsequent leachate). Significant effort has recently been invested in developing low alkali grouts which, whilst still carrying out their primary role of sealing water-conducting fractures, will degrade in a manner least likely to compromise long-term repository performance (see, for example, Emmelin et al., 2007, Karttunen and Raivio, 2008 and references therein). In general, it is expected that these grouts will behave as expected, but open questions remain regarding their competency following fracture re-activation (either due to tectonic events or glacial offloading)and potential freeze/thaw cycles potentially penetrating as deep as 300 m at the Olkiluoto site, for example (see discussion in Alexander and Neall, 2007).

Implications to repository performance

The quantitative implications of a hyperalkaline plume on the long-term performance of repository in crystalline rock have barely been examined. Most recent assessments of potential crystalline host rocks have either simply briefly mentioned the plume (e.g. ANDRA, 2005) or have assessed the implications for the EBS, but not the host rock (e.g. Pastina and Hellä, 2006) or simply not considered it at all (e.g. JAEA, 2007). In the last case, this is understandable due to the generic nature of the assessment.

Two examples exist where preliminary assessments of the implications of the hyperalkaline plume on specific repository sites have been carried out. In the first, Alexander and Mazurek (1996) considered the likely impact on the fractured marls of the Wellenberg site in Switzerland. Focus was on application of the data collected in the Jordan NA studies and the main conclusion was that it was necessary to conduct a detailed assessment of the likely long-term impact of gradual fracture sealing at the site due to the host rock reacting with the hyperalkaline fluids.

Vieno et al. (2003) assessed the likely impact on the EBS of hyperalkaline leachates (predominantly from fracture grouts) in the proposed Olkiluoto repository. The results suggested that little impact should be expected, but this was criticised by Alexander and Neall (2007) on the basis that many of the assumptions were unrealistic. Although Vieno et al. (2003) made use of hydrogeological data on the site, no attempt was made to assess the impact of the plume on the host rock itself and no other such studies have been reported to date.

Conclusions

Over two decades of work on various aspects of the reaction of (fractured) crystalline rocks to hyperalkaline fluids indicates that:

- the conceptual model for the evolution of a hyperalkaline plume in a crystalline host rock is largely consistent with observations from laboratory, URL and NA studies
- reactions between hyperalkaline waters and the host rock will modify the hydraulic conductivity of any fractures, significantly altering contaminent transport times and even the geometry of the flow field. Channelling effects could be significant, but this will vary from host rock to host rock
- most secondary reaction products have positive reaction volumes and thus fractures will be sealed by the precipitation of secondary phases
- (small aperature) fracture sealing occurs within short timescales (years to hundreds of years)
- the sequences of minerals predicted by coupled codes are generally very close to those observed in the hyperalkaline alteration zones, even if the specific phases cannot be represented due to a paucity of relevant thermodynamic and kinetic data

Generally, then, the phenomena associated with a hyperalkaline plume in crystalline rock can be effectively addressed by a combination of laboratory and field experiments supported by evidence from natural analogues. However, what remains clear is that the effects of the site hydrology (and tectonic/erosional processes) upon fracture sealing needs to be considered on a repository site-specific basis and this remains a challenge to PA modellers in the future. Additionally, the impact of fracture re-activation (due to tectonics or glacial unloading, for example) still needs to be considered.

References

Alexander, W.R. and Mazurek, M. (1996). The Maqarin natural analogue: possible implications for the performance of a cementitious repository at Wellenberg. Cited in Alexander and Smellie (1998).

Proc. NEA-IGSC Workshop on cementitious materials in safety cases for geological repositories for radioactive waste: role, evolution and interaction. Brussels, 17-20th November, 2009. Radioactive Waste Management, NEA/RWM/R(2012)3/REV, NEA/OECD, Paris, France.

W.R.Alexander and J.A.T.Smellie (1998) Maqarin natural analogue project. ANDRA, CEA, Nagra, Nirex and SKB synthesis report on Phases I, II and III. Nagra Project Report NPB98-08, Nagra, Wettingen, Switzerland.

Alexander, W.R. and Neall, F.B. (2007). Assessment of potential perturbations to Posiva's SF repository at Olkiluoto caused by construction and operation of the ONKALO facility. Posiva Working Report 2007-35, Posiva, Olkiluoto, Finland.

W.R.Alexander, C.A.Arcilla, I.G.McKinley, H.Kawamura, Y.Takahashi, K.Aoki and S.Miyoshi (2008). A new natural analogue study of the interaction of low-alkali cement leachates and the bentonite buffer. Sci. Basis Nucl. Waste Manag. XXXI, 493-500.

ANDRA (2005). Dossier 2005 Granite synthesis: Assets of granite formations for deep geological disposal. ANDRA Report, ANDRA, Paris, France.

Bateman, K., Coombs, P., Pearce, J., Noy, D.J. and Wetton, P. (1999). Fluid Rock Interactions in the Disturbed Zone: Nagra/Nirex/SKB Column Experiments - Phase II. BGS Technical Report WE/99/3C, BGS, Keyworth, UK.

Emmelin, A., Brantberger, M., Eriksson, M., Gustafson, G. And Stille, H. (2007). Rock grouting: Current competence and development for the final repository. SKB Report R-07-30, SKB, Stockholm, Sweden.

Haworth, A., Sharland, S.M., Tasker, P.W. and Tweed, C.J. (1987) Evolution of the groundwater chemistry around a nuclear waste repository. Sci Basis Nucl Waste Manag, XI, 645-651.

JAEA (2007). Second Progress Report on Research and Development for TRU Waste Disposal in Japan – Repository Design, Safety Assessment and Means of Implementation in the Generic Phase. JAEA-Review 2007-010, JAEA, Tokai, Japan.

Karttunen, P. and Raivio, P. (2008). R20 Programme: Grout Setting and Strength Development in ONKALO. Literature Review, Observations and Experiments. Posiva Working Report 2008-05, Posiva, Olkiluoto, Finland.

Mäder, U., Fierz, Th., Frieg, B., Eikenberg, J., Rüthi, M., Albinsson, Y., Möri, A., Ekberg, St. and Stille, P. (2006) Interaction of hyperalkaline fluid with fractured rock: Field and laboratory experiments of the HPF project (Grimsel Test Site, Switzerland). Journal of Geochemical Exploration, 90, 68-94.

Mazurek, M., Gautschi, A., Marschall, P., Alexander, W.R., Vigneron, G., Lebon P. and Delay, J. (2006). Transferability of features and processes from underground rock laboratories and natural analogues - Use for supporting the Swiss and French Safety Cases in argillacaeous formations. Proc. AMIGO II workshop, NEA, Paris, 2005. NEA/OECD, Paris, France.

A.E.Milodowski, C.A.Constantinou, W.R.Alexander, M.Rigas, C.Tweed, P.Sellin and P.Korkeakoski (2009). Reaction of bentonite in low-alkali cement leachates: preliminary results from the Cyprus Natural Analogue Project. Proc. 12th International Conference on Environmental Remediation and Radioactive Waste Management (ICEM '09), 11-15th October, 2009, Liverpool, UK. ASME, USA.

Pastina, B. and Hellä, P. (2006). Expected Evolution of a Spent Nuclear Fuel Repository at Olkiluoto. Posiva Report 2006-05, Posiva, Olkiluoto, Finland.

A.F.Pitty (*ed*), (2009). A natural analogue study of cement buffered, hyperalkaline groundwaters and their interaction with a repository host rock IV: an examination of the Khushaym Matruk (central Jordan) and Maqarin (northern Jordan) sites. NDA Technical Report, NDA, UK (*in press*).

Soler, J.M., Pfingsten, W., Paris, B., Mäder, U.K., Frieg, B., Neall, F., Källvenius, G., Yui, M., Yoshida, Y., Shi, P., Rochelle, C.A. and Noy, D.J. (2006). HPF experiment: modelling report. Nagra Technical Report NTB05-01, Nagra, Wettingen, Switzerland.

Vieno, T., Lehikoinen, J., Löfman, J., Nordman, H. and Mészáros, F. (2003). Assessment of disturbances caused by construction and operation of ONKALO. Posiva Report 2003-06, Posiva, Olkiluoto, Finland.