Cementitious colloids: integration of laboratory, natural analogue and in situ field data

## W.R.Alexander<sup>1</sup> and A.Moeri<sup>2</sup>

Goldschmidt Conference Abstracts, Geochim. Cosmochim. Acta, 18(S1): A11, 159-160.

1. Nagra, Hardstrasse 73, 5430 Wettingen, Switzerland. (<u>russell@nagra.ch</u> – now russell@bedrock-geosciences.com)

2. Geotechnical Institute, Gartenstrasse 13, 3001 Bern, Switzerland (andreas.moeri@geo-online.com)

## Abstract

In the case of a cementitious repository for radioactive or chemo-toxic wastes, it has been proposed that degradation of the cement may provide a significant source of colloids at both the cement/host rock interface and at the margins of the hyperalkaline plume produced by the leaching of the cementitious pore waters.

In the former case, five laboratory experiments examining colloid production during cement degradation and one natural analogue study at Maqarin in Jordan have been reported to date in the literature. Unfortunately, intercomparison of the data is difficult due to the significantly different methodologies used. Even in two batch leaching tests, large differences exist: in one, samples were shaken in synthetic cement pore waters while, in a second, static leaching in synthetic groundwaters was employed. The method-induced effects are apparent when the colloid populations are compared with the colloid mass concentrations: there are no clear relationships between populations and mass, suggesting significantly different colloid diameters are being measured in each experiment. This is clearly shown in the data of the first case where the measured colloid concentration varies over five orders of magnitude, depending on the settling time allowed following shaking of the samples (although the author notes that immediate measurement after end-over-end shaking of the batch cement/water samples used is unrealistic).

With respect to colloids produced at the margins of the hyperalkaline plume, no laboratory experiment has been able to reproduce this region so far and this margin has, as yet, not been accessed in the Maqarin study Arguably, the most appropriate approach to study such a margin would be in a large-scale, *in situ* experiment in an underground rock laboratory.

Clearly, any future work on these cementitious colloids would benefit from a common approach which should try to minimise method inherent differences, so producing a more compatible data set for use in assessing the likely impact of the colloids. In this paper, the existing cementitious colloid data sets are described in detail and a new, integrated approach for future work is defined. In addition, an *in situ* field experiment is proposed which will attempt to examine both the cement/host rock interface and hyperalkaline plume margin colloids simultaneously, so providing an integrated data set for impact assessment studies.

## References

- Alexander, W.R. and Smellie, J.A.T. (1998) Maqarin natural analogue project (synthesis report). Unpublished Nagra internal report, Nagra, Wettingen, Switzerland.
- Fujita, T., Sugiyama, D and Swanton, S.W. (2000) Investigations on colloid generation from cement hydrates and their characterisation. CRIEPI report U99057, CRIEPI, Abiko, Japan (*in Japanese*)
- Gardiner, M.P., Holtom, G.J. and Swanton, S.W. (1998). Influence of colloids, microbes and other perturbations on the near-field source term. The Analyst.
- Pearce, J.M. (1991). The changes in colloid population with time in laboratory microcosm experiments. BGS Report WE/91/2C, BGS, Keyworth, UK.

- Wetton, P.D., Pearce, J.M., Alexander, W.R., Milodowski, A.E., Reeder, s., Wragg, J. and Salameh, E. (1998). Production of colloids at the cement/host rock interface. Ch 10<u>in</u> J.A.T.Smellie (editor). Maqarin Natural Analogue Study: Phase III. SKB Technical Report 98-04. SKB Swedish Nuclear Fuel and Waste Management Co, Stockholm.
- Wieland, E. (1997). Colloid concentrations in cementitious backfill: monokorn morter and quartz in contact with hyperalkaline cement pore water. Unpublished internal technical note, PSI (Paul Scherrer Institute), Villigen, Switzerland.
- Wieland, E. and Spieler, P. (2001) Colloids in the mortar backfill of a cementitious repository for radioactive waste. Waste Manag 21, 511-523.

## Table 1 Comparison of existing data on near-field cementitious colloids (after Alexander and Smellie, 1998)

Report	Methodology	Colloid Population (mL <sup>-1</sup> )	Colloid Concentration (mgL <sup>-1</sup> )
Wetton et al., 1998	Collection of groundwater at the cement/host rock interface	1.15-4.89x10 <sup>7</sup>	0.051-0.190
Pearce, 1991	Incubation cells containing simulated waste, containment and backfill materials in synthetic groundwater	average 5x10 <sup>6</sup>	0.017ª
Wieland, 1997	Leaching of crushed PZHS <sup>b</sup> monocorn mortar and quartz aggregate with cement pore waters, solid:liquid ratio of 1:10	0.4-7.0x10 <sup>5</sup> (total) <sup>c</sup> 1.5-8.5x10 <sup>3</sup> (undist.) <sup>d</sup>	1-50 0.03-1
		$2-5x10^3$ (steady) <sup>e</sup>	0.004-0.021
Wieland and Spieler, 2001	Leaching of crushed mortar in a column with cement pore waters		0.002-0.058
Gardiner et al., 1998	Leaching of crushed cement with groundwaters, solid:liquid ratios of 1:5, 1:10 and 1:50	3-9x10 <sup>6</sup> (NRVB) <sup>f</sup> 1-2x10 <sup>5</sup> (PFA:OPC) <sup>g</sup> 1-9x10 <sup>5</sup> (BFS:OPC) <sup>h</sup>	As this paper is only an overview of unpublished data, full information is not available to calculate colloid mass

a. No data on colloid diameters available so smallest filter nominal pore size (15nm) used in the calculation. Note, however, that this calculation should be applied only to a size distribution and the error induced by using a single value for the colloid diameter may be up to two orders of magnitude (C.Degueldre, pers. comm. to WRA, 1996).

b. High Sulphate Resistance Portland Cement.

c. Total colloid concentration measured immediately after end-over-end mixing of a batch system. Experimentally defined colloid diameter ≥ 100nm.

d. Colloid concentration measured after leaving batch system undisturbed for 24 hours. Experimentally defined colloid diameters between 100nm and 1000nm

e. Colloid concentration measured after leaving batch system undisturbed for >24 hours. Experimentally defined colloid diameter ≥ 100nm.

f. Nirex Reference Vault Backfill. No data on colloid size distribution.

g. 3:1 mix of Pulverisied Fuel Ash: Ordinary Portland Cement. No data on colloid size distribution.

h. 3:1 mix of Blast Furnace Slag: Ordinary Portland Cement. No data on colloid size distribution.