Natural immobilization processes aid the understanding of long-term evolution of deep geological radioactive waste repositories

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The majority of readers of *Geochemistry: Exploration, Environment, Analysis* (GEEA) are doubtless well aware of the numerous geochemical studies of ore bodies around the world (e.g. GEEA 2005), and the natural interest that the nuclear power industry has in uranium ore bodies in particular as the ultimate source of fuel for nuclear power plants (NPPs). Perhaps fewer are aware, however, of the interest that the power industry also has in uranium ore bodies as indicators of long-term isolation of radioactive wastes ('radwastes') produced by the very same NPPs. Admittedly, this is a relatively young field of study, going back less than three decades to the pioneering work on the Oklo natural reactor site in The Gabon in West Africa and the Morro do Ferro Th–rare-earth element (REE) body in Brazil (see Table 1 for details).

The main thesis behind such studies is the very fact that many of these ore bodies have survived intact for aeons, despite significant changes in climate and geological conditions. They must have messages to impart which can be of use in the design and construction of radwaste repositories. This use of natural systems to help understand anthropogenic systems is generally termed 'natural analogues' (Chapman *et al.* 1984) in the radwaste industry. According to Miller *et al.* (2000), the term was first coined in the late 1970s and has since evolved. Côme & Chapman (1986) stated that a natural analogue was 'an occurrence of materials or processes which resemble those expected in a proposed geological waste repository'. McKinley (1989), however, stated that 'the essence of a natural analogue is the aspect of testing of models, whether conceptual or mathematical and not a particular attribute of the system itself', something which the IAEA agreed with when they stated (IAEA 1989) 'natural analogues are defined more by the methodology used to study and assess them than by any intrinsic physico-chemical properties they may possess'.

Table 1. *Examples of ore bodies studied as natural analogues of a radwaste repository (details, including significant references, can be found in Miller* et al. *2000)*

Site of ore body	Comments
Alligator River, Northern Territories, Australia	Shallow, secondary enriched uranium deposit
Broubster, Caithness, Scotland	Near-surface uranium mineralization
Cigar Lake, Saskatchewan, Canada	Deep, very rich[1] uranium ore body
El Berrocal, Toledo, Spain	Shallow, vein-hosted uranium ore body
Morro do Ferro, Minas Gerais, Brazil	Thorium–REE[2] ore body
Needles Eye, Solway Firth, Scotland	Near-surface, vein-hosted uranium mineralization
Oklo, Republic of Gabon	Shallow, uranium ore body[3]
Palmottu, Nummi-Pusula, Finland	Shallow, uranium–thorium ore body
Pena Blanca, Chihuahua, Mexico	Deep, uranium ore body[4]
Poços de Caldas, Minas Gerais, Brazil	Deep, uranium ore body[5]
Tono, Tokishi, Japan	Deep, uranium ore body[6]

[1] Uraninite and coffinite of average grade of 14% but reaching 55% in places (see also Gauthier-Lafaye *et al.* 2004).
[2] Up to 3 wt% Th and 20 wt% REE make this the most naturally radioactive place on the surface of the earth.
[3] The only known site of natural nuclear reactors, with a total of 16 identified to date. See Gauthier-Lafaye *et al.* (2004) for more details.
[4] The Nopal-1 mine.
[5] The Osamu Utsumi mine.
[6] The Tsukiyoshi ore body.

Despite these slight differences in the definition of natural analogues, one thing is certain: their unique role is to provide a link between the very short timescales of conventional field and laboratory studies and the enormous geological periods over which the results of such studies are extrapolated in radwaste repository performance assessments (PA). Further important roles attributed to analogues are their ability to access the true complexity/heterogeneity of real systems and the ease with which explanation by analogy is generally understood compared to the subtleties of PA mathematical models (see Fig. 1; Chapman & McKinley 1990; Alexander 1995; West *et al.* 2001). Over the last decade, indeed, natural analogue studies have been increasingly required by regulatory guidelines. Natural analogues are often presented as key components of national radwaste disposal programmes, one of the most extreme cases being in

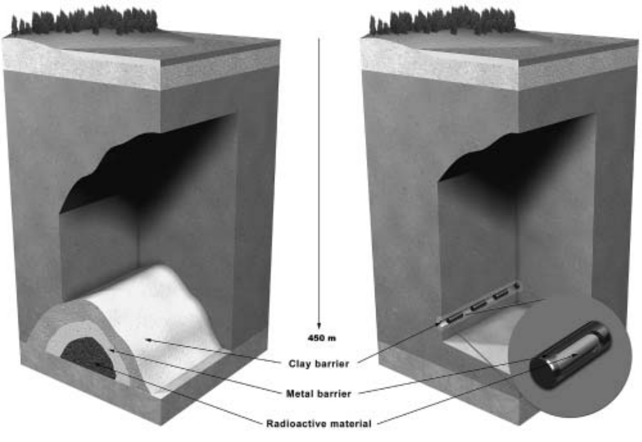

Fig. 1. Comparison of the Cigar Lake uranium ore body (left) with the engineered barrier system (right) for a repository for high-level radioactive waste (HLW). In Cigar Lake, the radioactive waste is represented by the natural uraninite ore (dark grey core), the metal barrier by an iron oxide/hydroxide rich zone (light grey middle layer) and the clay barrier by a clay-rich, hydrothermal halo (mid-grey outer layer). Image courtesy of DMmultimedia.

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Japan, where this requirement is explicitly stated in the nuclear law for the disposal of special wastes. It is fitting, therefore, that this special issue focuses on five natural analogue studies from Japan that were presented at a session (S34, Geochemical immobilization and long-term isolation of waste) of the Goldschmidt Conference at Kurashiki in Japan in September 2003.

The repository shown in Figure 1 clearly indicates the multi-barrier aspect of most current designs (see Witherspoon (2000) for details and other examples). In the engineered barrier system (EBS), the waste itself acts as a barrier to radionuclide release by being highly resistant to leaching. The thick steel overpack will corrode very slowly in the anoxic groundwaters of a deep repository and, finally, the thick, very dense, bentonite barrier material will preclude advective transport of any radionuclides to the surrounding host rock. Nevertheless, over geological time, the EBS is likely to degrade and those radionuclides that have not already decayed by this point will be released to the repository host rock, which acts as an additional barrier to radionuclide releases to the biosphere.

The papers presented here focus on this retardation (for retardation in the EBS, see examples in Miller *et al.* 2000) and use evidence from natural systems to assess how radionuclides released from the engineered barriers of a radwaste repository (Fig. 1, right) could be immobilized in the repository host rock. The five papers are divided into two main themes: natural retardation in the deep geosphere by Sasao *et al.*, Metcalfe *et al.* and Arthur *et al.*; and natural retardation at the geosphere/biosphere interface (GBI) by Akagawa *et al.* and Kanai *et al.*

In the first case, the papers present a broad overview of recent work on the Tsukiyoshi ore body at Tono in Japan. This research is of particular interest as this uranium ore body has experienced several cycles of uplift and erosion (with associated changes to the groundwater chemistry), a scenario which is relevant to many potential repository sites (e.g. in Finland, Sweden, and the UK). Further, the ore body is intersected by a major fault, a scenario of relevance to potential sites in tectonically active countries such as Japan, Taiwan and the USA.

Despite such significant geological upheavals, the Tsukiyoshi ore body persists and understanding the mechanisms behind this preservation is of direct relevance to establishing the long-term behaviour of many potential repository sites. In the second case, the papers examine radionuclide retention in near-surface rocks within the so-called 'GBI'. This is a very complex water–rock–biological interface zone with significant changes in pH and Eh of the groundwater system in a relatively confined rock volume, making modelling of radionuclide retention very difficult. For example, it has long been known that radionuclides may be retarded at near-surface redox fronts (e.g. Colley & Thompson 1985; Hofmann 1999), but this important aspect has generally been ignored when calculating the flux of radionuclides across the GBI and into the biosphere. Neglecting this process, of course, then tends to over-estimate likely doses to the surface environment, something which has become much more important in recent years as many PA modellers seek to make their dose calculations more realistic (e.g. Yoshida *et al.* 2006). Consequently, data such as those reported in these two papers will increase in relevance in the next few years and more studies of this nature can be expected.

The five papers here offer a short overview of ongoing work on natural analogues of geosphere radionuclide retardation in Japan. However, as noted above, the relevance of these studies is not limited to the Japanese radwaste programme; the information presented here will find a use worldwide.

The study based on the Tsukiyoshi ore body at Tono suggests that the deep geosphere can efficiently retard radionuclides, even when significant geological events disturb the site. Nevertheless, it is suggested that the work reported here be viewed as a preliminary understanding of the likely impact of such events and it is strongly recommended that further work be carried out at the site, focused on system understanding to make transferring the data collected here to a potential repository site more defensible.

The two papers on the GBI represent just a small portion of the work ongoing in this area today. It is likely that attaining a mechanistic understanding of the complex water–rock–microbial interaction processes in this zone will take time and it is recommended that a concerted, internationally co-ordinated, programme of work be carried out using broad-based (e.g. biologists, geochemists, hydrogeologists), well integrated teams. Only then is it likely that PA modellers will be able to represent the GBI more realistically in their calculations.

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