Nuclear waste disposal - the Oklo analogue

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Summary

Safe long term containment of anthropogenic nuclear waste is an urgent problem. Over the long term, geologic properties of specific sites together with various engineered barriers to contain nuclear waste, are the chief determinants of the total system. As natural analogues the Oklo natural nuclear fission reactors are unmatched. At Oklo, solid graphitized bitumen serves as the best of all barriers to the migration of ^{235}U and radiogenic nuclides. Similar engineered barriers therefore should be incorporated into nuclear waste depository designs.

Introduction

Safe disposal of radioactive wastes is one of the most urgent political and scientific problems facing society today (Ewing, 1999). Geological disposal, the favoured strategy, calls for deep long-term burial in a stable region in which the characteristic properties of the host rock promote isolation. Hazards of possible leakage are to be further reduced by various combinations of geologic and engineered barriers, including the most effective organic immobilization matrix, solid bitumen (Miller et al., 1994). The important task of correctly modelling actual geologic properties of a waste repository over the long term is a complex challenge that, according to some authorities, is currently being "downplayed" in the United States, where increased emphasis being placed on engineered barriers (Ewing, 1999). Choice of engineered barriers ranges from concrete, copper, glass, ceramics, steel and clay to bitumen. Of the latter two substances - both associated with the natural nuclear fission reactors of Oklo, Gabon, - bitumen more effectively constrains the migration of fission products (Nagy et al., 1991). Here we briefly review the role of organics in the world's best known natural analogues to a high level nuclear waste spill.

Geological Background

In Gabon, the Franceville series of sedimentary rocks lies unconformably on Archean crystalline basement. During the Paleoproterozoic, the coincidence of oxidized uranium-bearing solutions with hydrocarbon in common structural traps in these 2.1Ga old sediments, led to the deposition of uranium ores. Subsequently, pockets of high grade uranium ore went critical at Oklo and nearby at Bangombé, where today the remains of the only natural fission reactors known on Earth are located (Gauthier-Lafaye et al., 1996; Blanc et al., 1997). Uranium at Oklo was sourced from the basement Archean rocks, whereas liquid petroleum was primarily sourced during thermal maturation of syngenetic kerogen in organic matter-rich black shales near the base of the Franceville Series (Gauthier-Lafaye and Weber, 1989). Petroleum derivatives at Oklo are predominantly solid bitumen, which consists of condensed aromatic hydrocarbons interspersed with cryptocrystalline graphite Nagy et al., 1993).

In addition to locally depleting the ore in ^{235}U as a result of fission, the ore in reactor zones is characterized by the presence of fission products soluble in UO_2 (e.g., rare earth elements, Y, Nd and Zr, and various metallic or oxide inclusions such as Mo, Tc, Ru, Rh, Pd and Te). Together with hydrothermal clays and reactor zone phosphate minerals (notably hydroxyapatites), solid bitumen played a key role in constraining the mobilization and redistribution of uranium and various radiogenic nuclides (Gauthier-Lafaye et al., 1996; Nagy et al., 1983).

Exploring the natural analogue

According to the IAEA (1989) the most critical aspect of a natural analogue study is the manner in which it tests repository effects. For various reasons the Oklo natural reactors provide ideal subjects for study by workers concerned with the safe disposal of nuclear waste. For example, these reactors are analogues to modern pressurized water reactors in several significant respects: a) their cores consist of UO_2, similar to spent fuel, b) bitumen of Oklo reactors resembles the alpha-waste matrices proposed to confine wastes, c) hydrothermal clays surround them, similar to the clay backfill proposed for repositories, and d) fractured rocks surround the reactors as at many proposed repositories.

Our natural analogue research focusses on the geochemistry of solid bitumen associated with uranium ore at Oklo and Bangombé. Earlier studies employing reflected light microscopy, pyrolysis-gas chromatography-mass spectrometry and microfocussed laser Raman spectroscopy showed the solid bitumen to consist of a mixture of polycyclic aromatic hydrocarbons and cryptocrystalline graphite enclosing uraninite and entrapped fission-generated isotopes (Nagy et al., 1991; 1993). Thermal ionization mass spectrometry, x-ray fluorescence, neutron activation analysis and laser ablation-inductively coupled plasma-mass spectrometry (LAM-ICP-MS) (Nagy, et al., 1993; Jackson & Mossman, 1999) help confirm that at Oklo, the immobilization of uraninite in solidified graphitic bitumen enhanced containment of ^{235}U and various fission products. In this exercise, LAM-ICP-MS has proven most effective by providing *in situ* trace element and isotopic analyses of Oklo bitumen.

Preliminary studies are in progress to test the Oklo analogue at the Bangombé reactor (the so-called "last natural nuclear fission reactor on Earth" - Blanc et al., 1996), which lies buried in a deeply weathered tropical soil only 12 meters below the surface of the earth. The Bangombé reactor probably represents the best known natural analogue to the worst case scenario of a nuclear waste spill; specifically LAM-ICP-MS is being employed to evaluate the role played by its constituent solid bitumens as barriers to radionuclide migration.

Conclusions

The best natural analogue to a nuclear waste repository is one which supplies the basis of long term prediction. The Oklo fossil reactors are unique and valuable examples. The Bangombé reactor in particular represents a worst case scenario of a radioactive waste repository accident. Organic material in the form of solid bitumen has contributed to the preservation of this reactor, and as in the natural reactors at Oklo, this material may have served as an important barrier to the migration of ^{235}U and fissiogenic isotopes.

An engineered barrier of solid bitumen should therefore be incorporated into nuclear waste repository strategies.

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