NUCLEAR WASTE MANAGEMENT ORGANIZATION
SOCIÉTÉ DE GESTION DES DÉCHETS NUCLÉAIRES

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NWMO BACKGROUND PAPERS
4. SCIENCE AND ENVIRONMENT

4-3 NATURAL AND ANTHROPOGENIC ANALOGUES – INSIGHTS FOR MANAGEMENT OF SPENT FUEL

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NWMO Background Papers

NWMO has commissioned a series of background papers which present concepts and contextual information about the state of our knowledge on important topics related to the management of radioactive waste. The intent of these background papers is to provide input to defining possible approaches for the long-term management of used nuclear fuel and to contribute to an informed dialogue with the public and other stakeholders. The papers currently available are posted on NWMO's web site. Additional papers may be commissioned.

The topics of the background papers can be classified under the following broad headings:

1. **Guiding Concepts –** describe key concepts which can help guide an informed dialogue with the public and other stakeholders on the topic of radioactive waste management. They include perspectives on risk, security, the precautionary approach, adaptive management, traditional knowledge and sustainable development.
2. **Social and Ethical Dimensions** - provide perspectives on the social and ethical dimensions of radioactive waste management. They include background papers prepared for roundtable discussions.
3. **Health and Safety –** provide information on the status of relevant research, technologies, standards and procedures to reduce radiation and security risk associated with radioactive waste management.
4. **Science and Environment –** provide information on the current status of relevant research on ecosystem processes and environmental management issues. They include descriptions of the current efforts, as well as the status of research into our understanding of the biosphere and geosphere.
5. **Economic Factors** - provide insight into the economic factors and financial requirements for the long-term management of used nuclear fuel.
6. **Technical Methods** - provide general descriptions of the three methods for the long-term management of used nuclear fuel as defined in the NFWA, as well as other possible methods and related system requirements.
7. **Institutions and Governance** - outline the current relevant legal, administrative and institutional requirements that may be applicable to the long-term management of spent nuclear fuel in Canada, including legislation, regulations, guidelines, protocols, directives, policies and procedures of various jurisdictions.

EXECUTIVE SUMMARY

Canada has from the beginning of commercial power production in the early 1970s up until the end of 2002 produced just over 30,000 tonnes of spent fuel expressed as the uranium content. This spent fuel is now being stored at reactor sites with approximately 26,000 tonnes of uranium in wet storage and 4,000 tonnes in dry storage. With continued nuclear power production this inventory will continue to build in the future. The relatively small quantity of spent fuel that is produced from nuclear power generation means that adequate storage space at Canadian reactor sites could likely be made available to safely store spent fuel for decades to come.

Even though spent fuel from CANDU reactors contains considerable residual fuel value that could be recovered by reprocessing or reconditioning the fuel, this option is not currently being considered by Canada for a number of economic and political reasons. Accordingly, at this point in time, spent fuel from Canada's CANDU reactors is considered as a "waste" material that requires long-term management. The Canadian NWMO has been given the task of evaluating and consulting with the Canadian public on the options that are available for the long-term management of these "waste" materials. Generically these options entail either long-term storage near or at surface or deep geologic disposal. With the first option, monitoring, surveillance and recovery would be relatively simple, initial capital costs would be low but maintenance and surveillance costs would be high. In the second generic option, recovery would be difficult and capital costs high but ongoing surveillance and operational costs relatively low. A hybrid of these could also exist wherein the spent fuel could be "stored" for an indefinite time within a deep geological repository type of environment. This option would involve the highest level of all costs but also keep both disposal and recovery options open. The long-term management of spent nuclear fuel presents the same types of generic issues that society has been dealing with in the management of other types of hazardous waste. The general principles for management of hazardous wastes involve first trying to treat the waste so as to detoxify it. If this is not possible then the waste may be "treated and contained" such that it is prevented from entering the environment at concentrations that may be detrimental.

Strategies being developed globally for the long-term management of spent nuclear fuel recognize the fact that spent nuclear fuel becomes less dangerous with time as a consequence of the radioactive decay of its constituent elements. The first step in management of spent fuel is to contain it in underwater storage where, in the first ten years, approximately 99.9% of the radioactivity in the spent fuel removed from Canadian (CANDU) power reactors decays. At this point, a couple of management options are available.

- The spent fuel may be reprocessed during which the highly radioactive elements are separated from the less radioactive uranium. The less radioactive materials will then have to be managed for the long term and the uranium and other fissile materials recycled back into new fuel.

- The spent fuel for economic and/or social and political reasons is not reprocessed. This is the current situation for spent fuel from Canadian nuclear reactors.

If a decision is made not to reprocess the spent fuel management this initial wet storage period is followed by decades of dry storage where a further 90% of the radioactivity decays. Following the first 100 years after removal from the reactor in which 99.99% of the radioactivity has decayed, decay continues at a slower rate, such that the spent fuel will have decayed after a period of approximately 100,000 years to a radioactivity level (expressed as Bq/g of fuel) that is approaching that in natural uranium minerals such as uraninite (expressed as Bq/g) which were extracted from the earth and from which the fuel was fabricated in the first place. Once the radioactivity has decayed to this level, the remaining radioactivity of the spent fuel changes little over geological time, supported by the decay of the natural uranium, which constitutes over 98% the mass of the spent fuel. This suggests that any long-term management strategy should involve an initial period of secure containment of the order of tens of thousands of years and a longer term phase lasting over geological time (hundreds of thousands to millions of years) where the residual natural uranium constituents of the spent fuel will eventually be allowed to reintegrate back into natural global geochemical cycles.

Because of the long management time frames involved in the management of spent reactor fuels and the lack of any "engineering" precedence for the behaviour of containment systems designed to last over many thousands of years the argument is sometimes raised that the behaviour of spent fuel management systems cannot be reliably predicted. One can, however, look to natural and anthropogenic (man made) analogues to provide insight into how spent fuel waste management systems and their component parts may behave over these long time periods. In the natural and archaeological environments, there are analogues for spent fuel as it comes from the reactor as well as for spent fuel after it has decayed to activity levels found in natural uranium minerals. There are also analogues for the materials that may be used in engineered containment systems, such as cement, iron, copper and the clays proposed to contain spent fuel on surface or seal a deep geological disposal site. Analogues are also present that can provide insights into how various radionuclides behave as they cycle through the natural environment.

This paper provides a short discussion of a number of natural and archaeological analogues that can assist in understanding many of the issues associated with the long-term management of spent fuel. Analogues in the sense that they are used in this discussion paper do not "prove" that any particular concept is safe, but they can provide insights and serve to "bound" issues that may be of concern in carrying out safety assessments. In a similar manner, the fact that spent fuel after a period of time has the same total radioactivity as the original uranium minerals from which it was made does not mean that a deep geological repository will behave exactly the same as a natural uranium ore body.

Despite their limitations, analogues are useful, in that they can provide valuable insights and can serve to "bound" certain issues and concepts to be addressed in the management of spent fuel. In this context, analogues have been studied as a part of the nuclear waste management programs in many countries around the world.

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1.0 GENERAL CONCEPTS AND BACKGROUND

Canadian natural uranium deuterium reactors (CANDUs) burn natural (un-enriched) uranium enclosed in fuel bundles, with each bundle being about 0.5 m long and weighing approximately 20 kg. When the fuel can no longer be efficiently used for electrical power generation, due to fission product build-up, the used bundles are removed from the reactor core and replaced with fresh fuel. Spent fuel is predominantly uranium, and 98.7% of the uranium inventory present in fresh fuel remains present in the used or spent fuel. The remaining 1.3% has been converted to radioactive fission products, plutonium and small amounts of other transuranic elements as well as energy.

Most of the radioactivity present in spent fuel is associated with fission products, which generally decay rapidly to stable, non-radioactive forms. It is this radioactivity that presents the greatest potential hazard to human health and the environment in the first several thousands of years following the removal of spent fuel from the reactor. When removed from reactors, spent fuel is highly radioactive and is stored safely underwater on-site to allow thermal cooling and radioactive decay to occur in a shielded environment. After about ten years, only about 0.1% of the radioactivity found in fuel, as it is removed from the reactor, remains (Tait *et al*., 2000) and the spent fuel can then be stored safely on-site in specially designed dry-storage canisters within a dry storage facility. At CANDU nuclear sites, the spent fuel dry storage canisters have a nominal engineered design life of 50 years and an expected operational life of in excess of 100 years. Within 100 years, radioactivity levels in spent fuel will have decayed to about one ten-thousandth of the radioactivity originally present in the spent fuel (Tait *et al*., 2000). Thus, over time, the spent fuel continues to decay and pose less and less of a hazard until the radioactivity after about 100,000 years begins to approach the level of radioactivity associated with the decay products of natural uranium, as found in the original uranium minerals from which the fuel was derived. This is shown in Figure 1.

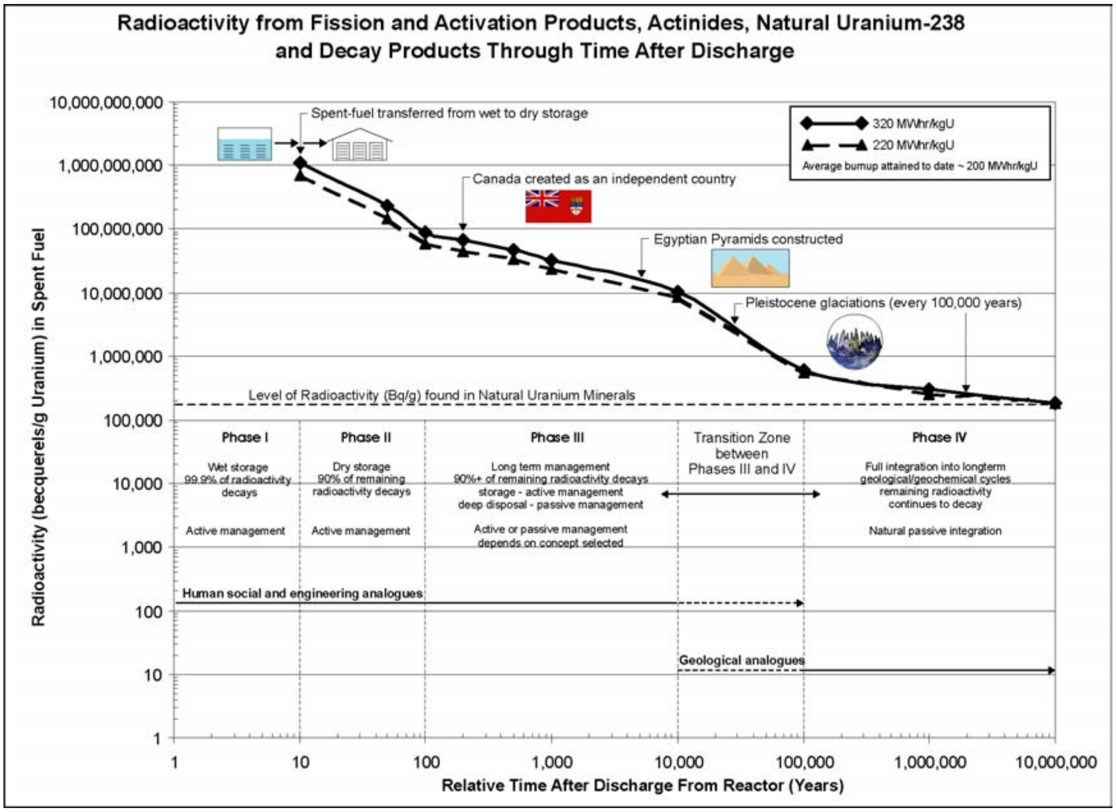

FIGURE 1: The level of total radioactivity remaining in spent fuel at various times following its removal from the initial 10-year wet storage period. Because fuel cycles and burnup efficiencies are continually changing, two curves are shown. The dotted line represents a burnup level in the spent fuel of 220 MWh/kg U and the solid line a burnup level of 320 MWh/kg U. The average burnup level in fuel from CANDU reactors as of the year 2000 was 200 MWh/kg U, so the data shown here may be considered as reasonable upper estimates of radioactivity likely to be present in spent CANDU fuel.

Using the simplified assumption that the degree of management required for spent fuel is proportionately to the level of radioactivity it contains, a series of four management phases could be envisioned corresponding to the activity levels shown in Figure 1.

The rate of decay of radioactivity suggests that containment or isolation of spent fuel from the surrounding environment is most critical in the initial 10 years after removal from the reactor when 99.9% of the radioactivity in the spent fuel decays. This is the time when spent fuel is presently being stored underwater for cooling and shielding at the reactor sites. During the next 100 years, a further 90% of the remaining radioactivity decays. During this period, the spent fuel is still very radioactive and requires good management and secure containment. Present plans envision aboveground dry storage at reactor sites for periods of on the order of 50 to 100 years. Over the next 100,000 years, a further 99% of the remaining radioactivity in the spent fuel will decay and, during this time, the spent fuel continues to remain more radioactive than the natural uranium minerals from which it was derived.

During this timeframe, beyond 100 years, the spent fuel should be managed in such a manner that releases to the biosphere are considered in any assessment. This is the timeframe during which society must evaluate options and make reasoned decisions on how best to design and implement systems for the “long-term” management of the spent fuel. Beyond 100,000 years, the level of radioactivity in a Canadian spent fuel management facility will be approaching that found in a uranium ore body and the same long-term geological/geochemical processes that are operational in nature will be responsible for integrating the residual materials back into natural geological/ geochemical cycles. There is not a sharp distinction between the end of Phase II and the beginning Phase III, and even much less of a distinction between the end of Phase III and the beginning of Phase IV, which is shown on the figure as a zone of transition.

It is assumed for the purposes of this discussion paper that the systems presently in place and licensed for wet and dry storage are appropriate for managing spent fuel for the first 50 to 100 years following its removal from the reactor. The period of need for decisions on long-term management thus extends from approximately 100 years following the removal of spent fuel from the reactor out to 10,000 years, or even 100,000 years at which time the level of radioactivity in the spent fuel approaches that of the natural uranium minerals from which it was derived and the total radioactive inventory of a facility containing all of Canada’s spent fuel is similar to the radioactive inventory of any one of a number of uranium mines in operation in Canada today

The basic generic options under consideration for the long-term management of spent fuel in Canada involve:

- an actively managed surface or near surface storage facility;
- a passively managed deep geological repository or possibly; and
- a hybrid concept of a deep geological storage facility from which spent fuel could be relatively easily retrieved or sealed into for disposal.

It should be noted that the overall radioactivity of the spent fuel is simply a gross indicator of the potential risk it may pose. Any safety analysis of a management concept will have to consider the specific radionuclides involved and their particular chemical, physical and radiological properties. For example, at 100,000 years following removal of the fuel from the reactor, the isotopes technicium-99 and plutonium-239 contribute more to the overall radioactivity than does uranium and its daughter products. Because their half-lives are significantly less than U-238, within a few hundred thousand years their contribution to residual radioactivity is much less, and uranium and its daughter products control the vast majority of the radioactivity in the spent fuel, as is the case in a uranium ore body. It is these element-specific chemical properties that may result in their mobilization from a management facility under a range of potential environmental conditions, and their subsequent behaviour in the geosphere and biosphere. It is this behaviour, which in turn results in the environmental risk they may pose and the overall safety of the system.

1.1 Is Spent Fuel a Waste?

In Canada, the government has committed to developing a policy and associated program for the long-term management of used nuclear fuel. Even though the spent fuel contains a significant quantity of uranium and other fissionable isotopes, it is presently not viable for a number of reasons in Canada to reprocess the fuel to recover these materials. If in the future Canada did move to a program of reprocessing spent fuel the reprocessing waste products would still require long-term management. As a rough first approximation the amount of radioactive waste to be managed over the long term until spent fuel has decayed to the equivalent of the level of radioactivity found in natural uranium minerals will be in direct proportion to the quantity of electricity produced. There is also research that suggests CANDU fuel with some level of reconditioning may be able to be used for a second re-burn cycle in light water reactors of the type being used in the USA. However, even if this or any other changes in fuel cycles were to occur, the spent fuel following a second “burn cycle” or future burn cycle will still require ultimate long-term management until the vast majority of the radioactivity has decayed away.

Accordingly, at present, spent fuel from CANDU reactors is considered a “waste material” that requires long-term management. The Nuclear Waste Management Organization (NWMO) is considering two potential generic long-term “management” concepts to manage this “waste”. The first concept is surface storage for an indefinite period of time. This concept of indefinite storage could be implemented at a national centralized facility or at one or more local facilities that may or may not be located at reactor sites. Indefinite storage could be implemented on surface or at various depths approaching and including that used for a deep geological repository. The facility would be designed for easy monitoring and retrieval and would require a high degree of active management and maintenance. The second generic concept is deep geological disposal, which would be designed in such a manner to blend passively over time into natural geological cycles and future retrieval would be difficult and expensive. Deep geological disposal could potentially occur at either a centralized national facility or in one or more local facilities that may or may not be located at present reactor sites.

1.2 Is the Management of Spent Fuel a Unique Problem?

The long-term management of spent nuclear fuel is often thought of as a complex technological and societal problem for which there is no precedent and for which, if the correct decision is not made, the consequences will be severe from both an economic, social and ecological standpoint. The issues associated with the long-term management of spent nuclear fuel are, however, very similar to the issues dealt with by society every day in managing dangerous waste materials of any kind, be they persistent toxins such as lead, mercury or cadmium, organic materials such as PCB waste or pesticides, or other radioactive materials such as mine tailings, medical wastes or scale from natural gas lines. The principles are generally the same, in that the first step in management is, if possible, to detoxify the waste so that it is rendered harmless. This works well with organic materials that can be incinerated to produce non-toxic water vapour and carbon dioxide. Short-lived radionuclides can, in a similar sense, be “detoxified” by isolating them and allowing them to decay to lower (in the case of spent nuclear fuel) or insignificant (in the case of some short lived medical isotopes) concentrations. Longer-lived radionuclides such as uranium and some less abundant fission products are similar to stable metals in that they have very long half-lives, and must be disposed of such that they slowly reintegrate over long geological times back into natural geological cycles. It is through the investigation of a number of natural occurrences of high concentrations of “natural” radionuclides in the environment and the study of their movement within the geosphere over long periods of time that one can gain insights into how spent fuel placed in a long-term management structure may also behave. It is perhaps worth noting that all radionuclides are “natural” in that they exist or have existed in the past in nature. It is simply that, in the production of nuclear power, short-lived radioactive materials are produced and will take time to again decay as they have in the past to very low trace concentrations.

The reduction in the potential radiation hazard associated with spent fuel over time as a consequence of radioactive decay can be illustrated by comparison of the total radioactivity inventory (expressed in Bq/g of uranium) present in spent fuel relative to the quantity present in natural uranium which is found in uranium ore deposits.

The comparison shown in Figure 1 provides a basis of reference for comparison with natural uranium deposits that can serve as “analogues” for deep geological storage/disposal of spent fuel. Thus, in a simple sense, the long-term management of spent CANDU fuel can be divided into two general time frames. During the first hundred years following the removal of spent fuel from a reactor, the radioactivity as measured in Becquerels (Bq)/g of uranium in fuel is very high and requires intensive active management. After a time period of approximately 100 years however, the radioactivity found in the spent fuel is much lower but still higher than that found in natural uranium minerals and an active or passive management system is required. Beyond 100,000 years a spent fuel management area will begin to take on the radiological characteristics of a uranium deposit and remain that way for billions of years.

1.3 Concepts for the Management of Spent Fuel

In Canada present day policy, economics and social norms all converge to assist in defining spent fuel as it is removed from CANDU nuclear power reactors as a waste material. These wastes must be managed, like other hazardous wastes for long periods of time. Although technologies exist for the recycling of spent fuel, these technologies are at present uneconomic and in conflict with current government policy and even if they were to be used in the future would still result in significant quantities of waste materials that would need to be managed over long time periods.

As discussed in the previous sections Canada is considering two generic approaches to long-term management. The first involves "long-term storage" of the spent fuel in one or more facilities located on surface or underground. The second generic concept involves "disposal" in a deep geological environment. In order for either of these concepts to be acceptable to the licensing authorities they must be able to demonstrate in a clear and transparent manner that the spent fuel managed by either of these concepts can be adequately contained so that no significant adverse effects occur in the surrounding environment. In order to ensure that this is the case the general philosophy employed elsewhere in the nuclear industry of "defense in depth" has also been employed by many national programs in the management of their spent fuel. This philosophy relies not on one key aspect of the management system for its success but rather on a number of interdependent systems such that if one fails the entire system is not put in jeopardy.

The application of this philosophy has generally resulted in systems for the long-term management of spent fuel that have a number of "barriers" that act in concert to prevent the release of materials to the surrounding environment. This is shown schematically in Figure 2.

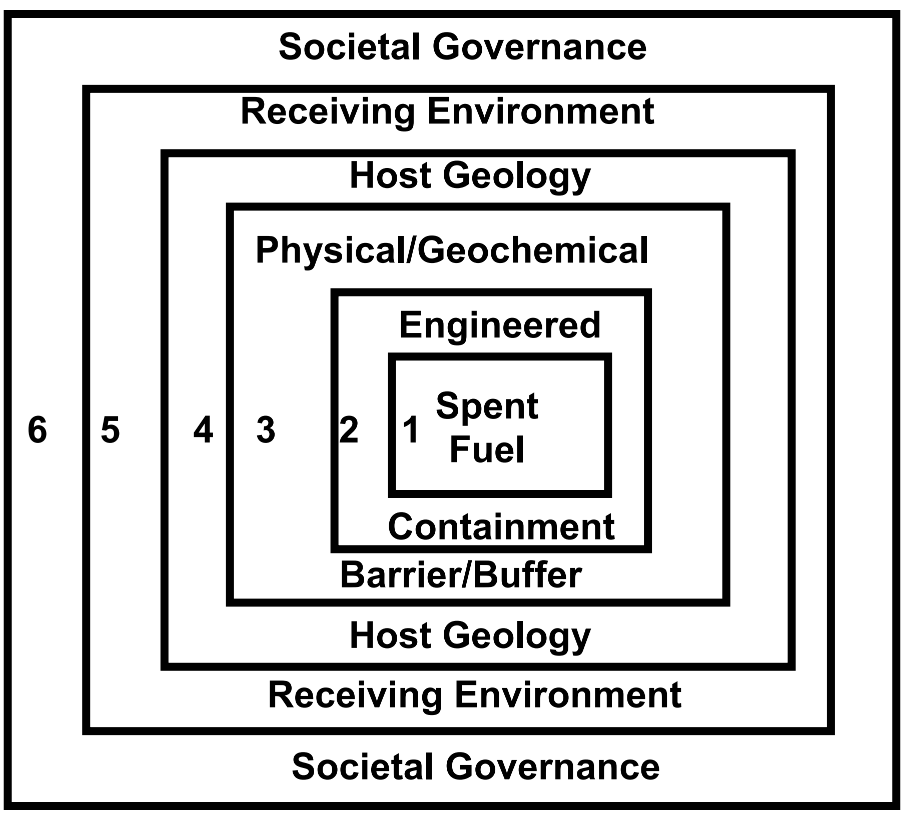

FIGURE 2: Illustration of the concept of defence in depth employed in the management of spent fuel.

In the above generic concept, the layers of defence include:

1. The spent fuel itself, which is comprised principally of ceramic uranium oxide fuel pellets that contain a number of radionuclides produced from the fission reactions in the reactor plus decay products of the uranium. The ceramic fuel pellets have been designed to have a very low solubility and provide a primary level of containment.

2. The fuel pellets are contained within a metallic fuel cladding, which is in turn packed with a buffer material to resist crushing and encased within a containment flask. The containment flask is manufactured of corrosion resistant materials and designed to last for long periods of time.

3. In the case of a deep geological disposal system the containment cask is in turn typically placed within a chemically resistant buffer material such as a swelling clay material that can slow or stop the movement of radionuclides or metals elements through it. In the case of a long-term storage facility the cask may be placed within a large cement dry storage cask on surface or a similarly secure containment structure underground.

4. In the case of a deep geological disposal facility, the natural geology provides the final barrier between the materials contained in the spent fuel and the natural environment. In the case of a surface or near surface facility, an engineered building or other structure would provide this barrier.

5. The next barrier is the natural environment. A “good” natural environment barrier in this context is one where there is a high degree of dilution and the absence of a “sensitive” ecosystem.

6. The final barrier could be considered societal governance will provide for long-term surveillance, protection and repair of the various barrier systems as is necessary.

The essential difference between the concept of long-term storage and deep geological disposal is that:

1. In the case of long-term surface storage, societal governance (barrier 6) is relied on to ensure long-term security and maintenance of the initial containing systems (barrier 3 in the case of a deep subsurface storage location and barriers 3 and 4 in the case of a surface storage system)

2. In the case of deep geological disposal, natural geological and geochemical processes are relied upon to ensure the proper functioning of the secondary containment and natural geology (barriers 3 and 4). Since deep geological disposal is designed to be a passive management system it does not rely on long term societal governance.

This paper discusses a number of various analogues for each of the types of barrier components that may be designed into a long-term spent fuel management concept.

2.0 ANALOGUE CONCEPT

The various concepts that may be employed in the long-term management of spent fuel can benefit significantly from the study of natural and anthropogenic analogues for processes expected to occur at management sites. Analogues are features found in the natural or man-made environment that provide understanding of how a radioactive waste management facility may behave over time scales ranging from thousands to perhaps millions of years. Natural analogue studies can inform scientists and engineers about how to design better facilities, and can contribute to the assurance of the general public and nuclear regulatory agencies that management facilities can be designed and operated safely over the long term. The study of natural analogues has application in principle to both the long-term storage and the deep geological disposal concepts.

In the case of long-term surface storage, analogues could potentially consist of surface or near surface structures designed by human civilizations to remain in an undisturbed state for long periods of time. The most commonly cited analogues in this category are large religious structures such as the Egyptian and Central and South American pyramids, or protective structures such as the Great Wall of China or Hadrian's Wall in Britain, all which have been in existence for many hundreds or thousands of years. These examples can provide insights concerning the ability of man-made structures and the materials they are constructed from to physically last over long time periods with little subsequent maintenance. They can also provide insights into how different human societies and cultures may treat and view these types of structures over similarly long periods of time. The focus of this present paper, however, is not on the stability of the social institutions, which were responsible for the construction of these structures, but rather on the many scientific and engineering insights we may be able to obtain from these structures and the materials they contain.

In the case of deep geological storage/disposal, there are also a number of "natural" analogues that can provide insights into the long-term behaviour of spent fuel, should it be managed by deep geological storage or disposal.

2.1 Natural Analogues for Deep Geological Disposal

Natural analogues for waste repositories have attracted strong interest in recent years (IAEA, 1989, 1999, 2003; CEC, 1987; EC, 2000; Miller *et al.*, 2000; Murphy, 2000; Smellie *et al.*, 2000). Major review texts on the subject were published by Alexander *et al.* (1994) and Miller *et al.* (2000), which cite a great volume of literature on the subject. A major international conference called "Natural and Man-Made Repositories - Paradigm for Nuclear Waste Policy" was held recently (5-7 November 2003) in Bucharest.

Natural analogues are used in many cases to provide models for specific components or processes associated with the design and evaluation of waste repository concepts, and to support or complement laboratory or field-based measurements. These types of study can be factored into performance assessment models for simulation of physical and chemical processes at spent fuel repositories. Natural analogue studies can be used to obtain quantitative or semi-quantitative information on the behaviour of radionuclides within specific types of geological environments and engineered containment systems. Beyond their application in performance assessment, natural analogues serve to illustrate natural parallels for the key features associated with spent fuel repositories. As such, parallels can be used to provide strong, real-world evidence that can be used by policy-makers, politicians, academics and the public at large to provide insights into the safety of any proposed disposal or storage concept. As such, documentation of appropriate natural analogues and their behaviour over long periods of time provides another piece of supporting evidence for safety assessments of deep geological disposal or storage concepts. Before any safety assessment can be judged as credible by the regulatory authorities and the Canadian public several independent lines of evidence will be necessary in supporting any concept that is put forth. Natural analogues exist for many components of a waste repository, including analogues for the behaviour of spent fuel itself, for engineered and natural containment systems or barriers that may be used to inhibit contaminant migration, and for transport processes that could potentially allow contaminants to migrate to the environment. This background paper provides a few illustrations of each type of natural analogue, and outlines the key information arising from them that can provide insights that may assist in the design and implementation of a deep geological disposal or surface storage concept and in the building of credibility for any concept that may ultimately be proposed.

3.0 ANALOGUES FOR SPENT FUEL

Natural uranium (predominantly a mixture of U-235 (0.7%) and U-238 (99.3%)) and natural thorium (predominantly Th-232) are relatively abundant radioactive elements on earth. Like all other elements, they are cycled through biological and geological systems and tend to be concentrated and again dispersed by natural processes. Uranium, for example, will slowly dissolve under oxidizing conditions and precipitate in the absence of oxygen under reducing conditions. It is this basic chemistry that causes most uranium ore bodies to form and remain in place over many hundreds of millions of years. Ore bodies that are being mined today were formed hundreds of millions of years ago. Other materials, such as many coal deposits, contain elevated concentrations of uranium that result from very dilute concentrations of uranium being carried by groundwater, leached from rock such as granite, percolating through the reducing environment in the peat that eventually turned into coal. In some areas, the concentrations in coal have risen to levels such that recovery of uranium from coal ash has become economic. This same process is ongoing today when dilute solutions of uranium in groundwater or surface waters encounter chemically reducing conditions causing the uranium to precipitate, such as in some peat deposits. In these environments, groundwaters deposit the uranium they carry into the reducing environment of the peat, elevating the uranium concentrations in the peat. Thorium on the other hand has a different chemistry and readily reacts in the earth's crust to form a dense, hard, chemically stable phosphate called monazite. This mineral weathers from rock and can accumulate in relatively high concentrations in placer deposits. Although natural thorium (Th-232) can be used in a reactor fuel cycle, thorium reactors are not in commercial use. Accordingly, this discussion will focus on analogues associated with uranium, which is the dominant nuclear fuel used in commercial reactors today. Thorium is of interest, however, both as a chemical analogue for plutonium, an important component in spent fuel, and because thorium-230, a decay product of uranium, is also present in spent fuel..

Spent fuel consists predominantly of uranium dioxide (UO_2) along with fission and activation products resulting from the nuclear reactions occurring in the fuel during power production. Natural uranium minerals in many ore bodies consist of UO_2 along with uranium decay products. In some other ore bodies where the conditions were right and the concentration of U-235 (the isotope of uranium capable of sustaining a nuclear fission reaction) was high enough to sustain a chain reaction, depleted uranium (with a low U-235 content) is also present along with some long-lived fission products and the normal uranium decay products. Studies of both types of uranium ore bodies can provide valuable insights into the long-term behaviour of spent fuel.

In a natural uranium ore deposit there are "pockets" of minerals such as uraninite (UO_2), the quantity of which relative to other minerals in the mined material dictates the grade of the material. In some "rich" ore bodies such as the Cigar Lake deposit discussed elsewhere in this paper, there are "pods" of ore of significant size with in excess of 50% uranium. This uranium ore consists of the natural uranium parent isotopes U-238 and U-235 plus their decay products in “secular” equilibrium with their parents. Natural uranium consists of approximately 99.27% uranium-238 and 0.71% of uranium-235. A very small mass of uranium-234 is also present. Normally, uranium as it is mined from ore is present in equilibrium with all of the natural decay products of these two dominant isotopes, including isotopes of radium, thorium, radon, bismuth, polonium, lead etc. This radioactivity inventory will only decrease very slowly with time, controlled by the half-life (4.5 billion years) of the predominant parent radionuclide U-238.

Accordingly, one gram of natural uranium, as it is extracted from the earth in equilibrium with all of its daughter products, contains about 178 thousand becquerels of radioactivity. In comparison, after discharge from a reactor and 10 years of cooling and decay underwater, spent fuel has an inventory of 750 million becquerels of radioactivity per gram of uranium in the fuel, principally in the form of fission and activation products (Tait *et al.,* 2000), and is more than 40 thousand times more radioactive than is the original uranium ore. Due to the rapid decay of most of the fission product isotopes present in spent fuel, after approximately 100,000 years, spent fuel has decayed to a similar level of overall radioactivity as found in the uranium mineral uraninite used to produce the fuel in the first place (Figure 1).

As discussed in Section 1.0, spent fuel in terms of its total radioactivity can be considered conceptually to have four periods of management. The first two would be for the first hundred or so years of active management following removal from the reactor when the great majority of the radioactive decay occurs. The third would be for the following hundred thousand years or so until the ingrowth of uranium decay products begins to dominate the radioactivity in the spent fuel, as it begins to take on the radioactive characteristics of rich uranium ore bodies such as those presently being mined in Canada. It is this third period which is considered as necessary for long-term management. Beyond this timeframe, the level of radioactivity and the risk posed by the spent fuel is similar to that of a rich uranium ore body located in a similar type of environment.

Standard engineering designs are accepted, proven and licensed for spent fuel in both wet (first 10 years or so) and dry (estimated design life of approximately 100 years) storage containment structures as are presently in use at CANDU reactor sites. It is for the time periods beyond the first 100 years of management that analogues can provide us valuable insights.

There are two general types of uranium ore bodies that can provide valuable analogue insights to deep geological disposal of spent fuel. The most common type of uranium ore body contains uranium in the ratio of 99.3% U-238 and 0.7% U-235. The second and much less common type of ore body is depleted in U-235 (containing less than 0.7% U-235) as a consequence of natural fission reactions that have occurred in the ore body in the past.

The investigation of these two types of natural uranium ore bodies can thus give valuable insights into the long-term behaviour of spent fuel of various ages within a number of specific geological environments where these ore bodies are located. The following provides a short discussion of examples of these two types of ore bodies. The first is a "depleted" ore body in which the concentration of U-235 is below the expected value of 0.7% as a consequence of its fissioning in the past. The second is an undepleted ore body containing uranium in its normal isotopic abundances.

3.1 Depleted Uranium Orebody - The Oklo "Reactors"

It generally comes as a surprise to most members of the general public that man did not invent the nuclear fission reactor but rather just rediscovered what has occurred naturally as uranium fission reactors over 2 billion years ago. It is the remnants of these natural uranium fission reactors which provide the closest natural analogue for the disposal of spent fuel over long time periods in a geologic environment. There are 15 such "fossil" reactors known in Gabon, Africa. An excellent overview of the Oklo reactors in Gabon is provided by Dr. R. Loss at: www.curtin.edu.au/curtin/centre/waisrc/oklo/index.shtml.

The fission reaction occurs when U-235 absorbs a small sub-atomic particle like a neutron, causing it to split into fission products and more neutrons. A fission reactor requires a high overall concentration of uranium, specifically the uranium isotope U-235, which is fissionable, a low concentration of neutron absorbers which can "poison" the reaction, and a high concentration of a moderator which slows the neutrons, preventing a runaway reaction such as occurs in a nuclear explosion. The ejected neutrons can be absorbed by neighbouring fissionable U-235 nuclei, leading to a sustained chain reaction.

The quantity of U-235 present in natural uranium ore bodies today is low at 0.7%. However, approximately 2 billion years ago, when the Oklo ore bodies formed, the fraction of U-235 present in natural uranium was much greater, comprising just over 5% (less had undergone natural decay). At Oklo, concentrations of uranium in ore body were also very high (up to 70% uraninite comprised of uranium dioxide) and this coupled with the high (relative to today) isotopic concentration of U-235 resulted in conditions, which could sustain the nuclear fission reaction. This uranium concentration was also coupled with low concentrations of trace elements in the ore body such as boron or vanadium, which if present could have poisoned the fission reaction and prevented it from continuing. At Oklo the water and carbon in the deposit provided a moderator, which slowed neutrons so that they interacted with other U-235 nuclei and caused the reaction to be sustained. These factors, combined with a sufficient mass of uranium within individual pockets, produced natural fission reactors, the remnants of which we see today. In comparison with the natural reactors that operated at Oklo, CANDU reactors use heavy water to moderate the fission reaction and use neutron absorbers such as certain metals to poison the reaction if a shutdown is required.

Radioactive "clocks" (very small concentrations of fission products found in the Oklo reactors that could only have been produced in a fission reactor) have been used to determine that these natural nuclear reactors operated at low power over about 1 million years to some 2 billion years ago. Evidence also shows that some 6 to 12 tonnes of U-235 underwent fission, producing some 4 tonnes of plutonium and generating temperatures in the natural reactors of up to 600°C.

The Oklo reactors are not perfect analogues for the geological disposal of spent fuel from power reactors. This is because the Oklo "spent fuel" was generated and "disposed of" within the natural reactor core, and was not encased in metal cladding or surrounded by engineered barriers. The Oklo "reactors" also produced lower concentrations of fission products than found in modern day spent fuel, had lower operating temperatures and had much longer operating "lives" than a modern power reactor. Nevertheless, Oklo reactor studies have provided important information on the stability and longevity of uranium dioxide (uraninite) in the presence of other fission products, the transport and retardation of radionuclides within the surrounding geological environment, degradation and hydrolysis of bitumen formed by organic matter adjacent to the reactors, and the production of radiologic oxidants. All of these factors are very important considerations in the consideration of any concept involving deep geological disposal of spent fuel. Studies at Oklo have shown that more than 90% of the uraninite "fuel" present in the reactors 2 billion years ago has remained in place, as have the transuranic elements and many of the fission products and their decay products, despite the absence of "engineered" barriers that would be required for a geological repository today. This has been the case despite two billion years of continental drift, groundwater movement in the area, and despite the present day near-surface location of these natural reactors. Oklo has also shown us that some long-lived fission products such as iodine-129 did migrate from the "spent fuel" in these reactors emphasizing that we must pay careful attention to these more mobile elements in any safety assessment of spent fuel.

As illustrated in Figure 2, plants and animals thrive at the exposed faces of these natural fuel repositories where the levels of radioactivity have long ago decayed to those typically found adjacent to a uranium ore deposit (which they now are). Over time, spent fuel eventually becomes relatively low in radioactivity, with a much reduced potential for causing harmful effects in the surrounding environment.

What the Oklo "fossil" reactors allow us to see is a snapshot in time of the condition of a natural spent fuel repository two billion years after "decommissioning". Without the engineered barriers around it, Oklo provides what might be considered a worst-case representation. What these fossil reactors do not allow us to see directly is their behaviour following their "decommissioning" up until the present time. This information has to be gleaned only from indirect evidence such as the quantity of fission products and their decay products, and actinides that can still be found in association with these "fossil reactors". It is this indirect evidence that suggests that the mobility of many fission products and actinides from a modern day spent fuel geological repository is likely to be low in certain geological environments, as it was in the past.

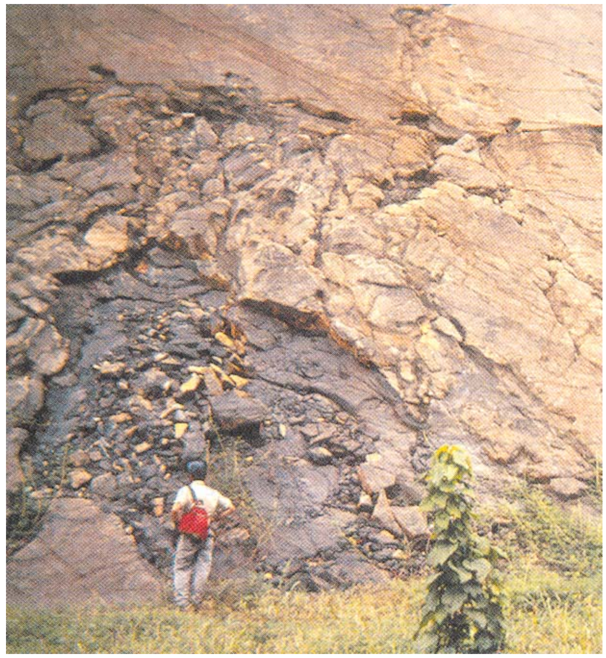

FIGURE 2: Figure 2 shows an exposed outcrop in Gabon Africa in which the remnants of a two billion year old natural nuclear uranium fission reactor can be seen. When the reactor “shut down” after approximately 1 million years of operation it contained all of the fission products that would be found in spent fuel. Traces of some of the longer-lived fission products and the daughter products of the shorter-lived fission products can be still found in the reactor along with uranium and its daughter products, which today dominate the radioactivity in the deposit (from Miller *et al*., 2000, reproduced with permission from Elsevier Science)

3.2 Natural Uranium Deposits - Cigar Lake

Unlike the Oklo fossil reactors, the Cigar Lake uranium deposit found in Northern Saskatchewan did not become a natural reactor. The ore body formed more recently and perhaps did not have the proper combination of sufficient percentage of U-235, neutron moderators and geometry to allow it to undergo fission. Nevertheless, the ore body provides an excellent Canadian example of an analogue for geological disposal of spent fuel. The Cigar Lake deposit is currently being developed as a uranium mine. The Cigar Lake deposit has been well studied as a natural analogue (e.g., Cramer and Smellie, 1994; Miller *et al.*, 2000).

The Cigar Lake uranium deposit is currently located at a depth applicable to deep geologic disposal, about 430 m below surface. Uranium is present predominantly as uraninite (UO_2), as in spent fuel, and as coffinite, with an average economic grade of about 8% and a maximum of greater than 60% in some areas. The total uranium reserves in the ore deposit are approximately 150,000 tonnes. To put this into perspective, the total quantity of uranium in spent fuel in Canada is approximately 30,000 tonnes or less than 20% of that which is present in the Cigar Lake ore body. According to the IAEA the total quantity of spent fuel in storage on a global basis is 171,000 tonnes expressed as U.

The Cigar Lake ore body formed about 1.3 billion years ago so that the percentage of U-235 present in the uranium was less than at Oklo at its formation some 700 million years earlier. This lower abundance of U-235 (estimated at 2.8%) at the time of formation of the Cigar Lake ore body, plus the presence of neutron-absorbing elements and insufficient quantities of a suitable moderator, combined to prevent a sustained nuclear chain reaction from occurring at Cigar Lake. Nevertheless, the uraninite mineral present is comparable to the uranium dioxide, which constitutes more than 98% of the mass of spent fuel. The UO_2 that constitutes the majority of the mass of spent fuel is different from the UO_2 in the uranium mineral uraninite. The UO_2 in spent fuel contains within the fuel matrix a number of fission products and actinides that endow it with a much higher level of radioactivity than the uraninite mineral found in uranium ore for a few hundred thousand years. In a very simplified sense the Cigar Lake ore body could be thought of as a single repository containing the same quantity of radioactivity as a repository containing all of the spent fuel produced in the world up until the end of 2002 following a subsequent period of decay of several hundred thousand years.

The Cigar Lake deposit has a similar depth of host rock and geometry to possible repository designs, the ore is similar in general composition to spent fuel, and the ore is surrounded by a clay envelop somewhat analogous to a clay buffer that could be placed around spent fuel in a deep geological repository. The ore itself is not, however, surrounded by metal containment, as in the case of spent fuel and the ore does not contain the fission and activation products resulting from the presence of a chain reaction, as was the case at Oklo. It can be considered analogous to a “worst-case” repository design, as it lacks any specially designed engineered barriers, and the host rock above the ore body is highly fractured sandstone (see Figure 3).

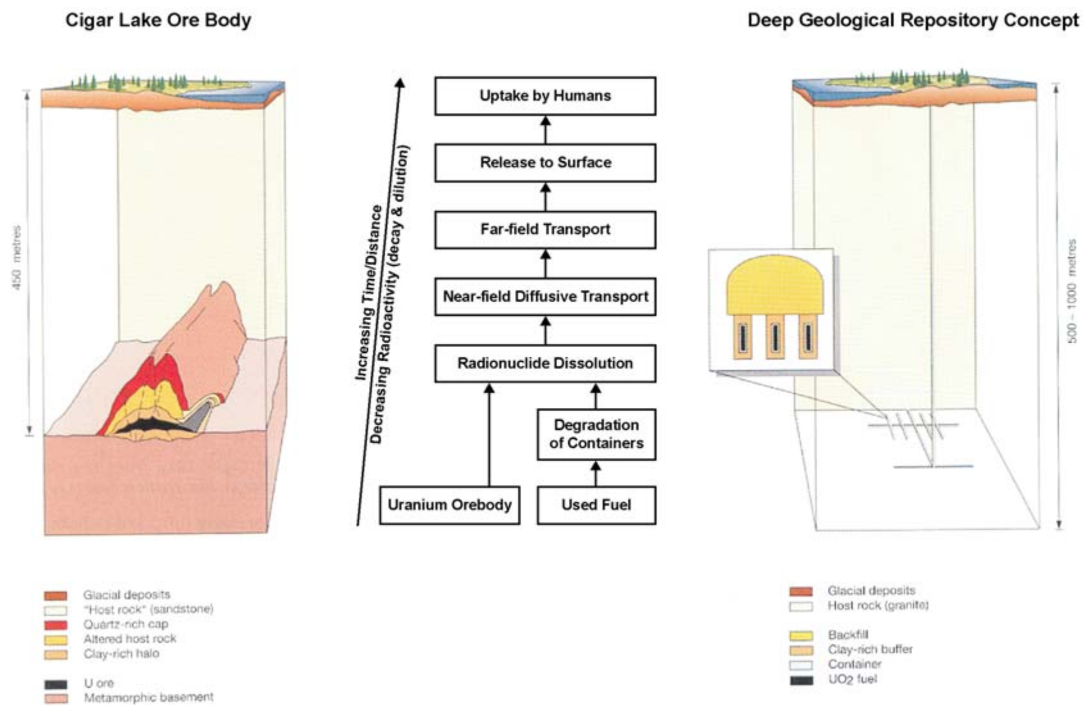

FIGURE 3: A schematic of the Cigar Lake ore body is shown on the left side and a schematic of a spent fuel geological management system is shown on the right. The center shows the various processes that have in the past influenced the transport of radionuclides from the ore body and the same processes that will in the future influence the transport of contaminants from a deep geological repository (adapted from Miller *et al*., 2000, reproduced with permission from Elsevier Science)

Based on the Cigar Lake natural analogue study (Cramer and Smellie, 1994), scientists have concluded that:

- the uraninite mineral containing the UO_2 is stable under the chemically-reducing conditions found adjacent to the Cigar Lake ore over 100 million year time scales, with very little uranium dissolving in groundwater moving significantly from the deposit;
- the natural clay surrounding the ore has provided an effective long-term seal, preventing movement of radionuclides away from the deposit and preventing movement of colloids and radionuclides they may transport through the deposit;
- dissolved organic matter in groundwater migrating past the ore has not played a significant role in mobilizing radionuclides from the deposit; and
- natural hydrologic barriers and appropriate geochemical conditions found at the site are effective in preventing significant radionuclide migration from the deposit.

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Insufficient radionuclide migration has occurred from the Cigar Lake deposit to produce any detectible concentration anomalies in the soil, surface water and lake sediments and waters overlying the ore body. Environmental and geological exploration in the area has shown no surface expression of the ore body, and it had to be discovered by geophysical techniques applied at depth. Indeed, on a map of surface radioactivity in Canada, the area of the Saskatchewan deposits generally shows up as having below-average surface radioactivity.

The Cigar Lake analogue study provides a large volume of data on prevailing physical, chemical and biological processes, which can provide important information to be used in the siting and designing of deep underground repositories for spent fuel. Many repository designs proposed for the disposal of spent fuel have a general design similar to the ore body found at Cigar Lake, in which the uranium and radionuclides found in association with it are encased in a geochemical buffer system composed of clay. Although the Cigar Lake site has been the most extensively studied site in Canada as a geological analogue for a deep geological repository, there are several other large and rich uranium ore bodies found in both similar (McArthur River, McClean Lake, Midwest Lake) and different geological environments (Cluff Lake, Rabbit Lake, Beaverlodge, Gunnar, etc.) in Saskatchewan that provide insights into the fact that large concentrations and quantities of uranium and its decay products can remain stable (both physically and chemically) in a range of geological environments over very long time periods (billions of years). Under some conditions however, uranium can be mobile over long time periods.

3.3 Roll-Front Uranium Deposits

In some parts of the world, uranium found in permeable rocks is mined by “in situ” mining methods. Some of the ore bodies mined in this manner are called roll front ore bodies, as they are continuously migrating or rolling through the permeable host rock such as sandstone. The front of the ore body is in a reduced state, while the rear of the ore body is in a more oxidized state as a consequence of oxidizing groundwaters that are “pushing” the ore body along through the rock formation in geological time. This creates a condition at the rear of the ore body in which the uranium becomes soluble and migrates to the front of the ore body where it again precipitates when it encounters reducing conditions. A well-studied example of a roll-front uranium deposit is the Osamu Utsumi mine in Brazil (Hoffmann, 1999). By investigating the materials such as iron or organic carbon that are responsible for creating certain oxidizing or reducing conditions, we can understand how uranium and associated radionuclides in spent fuel can be mobilized in a waste disposal facility and alternatively the conditions that will result in their precipitation and consequent immobilization.

3.4 Other Deposits

There are many known uranium deposits in the world, and most could similarly be studied as analogues for spent fuel disposal in geologic environments. Among these, the Peňa Blanca ore body in Mexico located well above the water table has been studied as a good analogue for the Yucca Mountain repository in Nevada. The Alligator River deposit in Australia and the Pocos de Caldas deposit in Brazil have been studied in the context of uranium migration behaviour in shallow more tropical and very high rainfall locales.

Uraniferous peat and coal deposits provide geologically more recent evidence of the geochemical mobilization of uranium under oxidizing conditions in the source rock and its subsequent precipitation of immobilization in the peat or coal.

Thorium provides a chemical analogue for plutonium behaviour in its solid state (Pu^{+4}), and several plutonium analogue studies have been completed at Morro do Ferro, Brazil (Miller *et al.*, 2000). The geochemistry of plutonium is however complex as it is capable of existing in a number of oxidation states under a range of geochemical conditions ($Pu +^{2,+3,+4,+5}$) and which can exhibit different degrees of chemical reactivity and mobility in the environment. This again emphasizes the need for being able to design and build a long-term management system for spent fuel which provides a high degree of long-term physical and geochemical stability that will last several hundred thousands of years.

4.0 ANALOGUES FOR BARRIERS

Spent fuel, whether managed for the long term at surface or deep underground, will have engineered barriers designed to strongly inhibit or prevent movement of radioactive elements and other materials from the facility into the surrounding environment. Movement of radionuclides can be prevented by the use of both physical and geochemical barriers. Physical barriers are generally designed to prevent the movement of groundwater, particulates or gas into or out of a repository. Geochemical barriers may allow the movement of groundwater or gas but inhibit the movement of radionuclides or some stable trace metals through sorption or precipitation reactions. In practice these types of barriers are often combined such as the use of iron as both a physical containment system and a redox barrier that will tie up oxygen as oxides of iron (rust), preventing oxidation and possible dissolution of uranium and adsorbing other elements. In the same manner natural materials such as swelling clays can be used to provide watertight barriers and serve as good chemical sorbants for radionuclides, further slowing any diffusion that may occur through them. Other barrier materials such as copper may be selected for their physical containment properties as well as their thermodynamic stability (meaning that they are chemically non reactive) within the geological environment of the repository.

Typical designs use a multi-barrier concept, which includes materials such as alloys of iron or other metals, concrete and clays such as bentonite. A generic design used in underground disposal would typically include the waste, which is contained in Zircaloy fuel rods, packed and immobilized in a drum or canister, with an immobilizing filler. These containers would typically be packed in larger containers and placed within an underground vault. The vault itself might be filled, as discussed above, with an immobilizing and swelling material such as a swelling clay around the containers. The repository would be sited at a location that affords good natural containment, owing to the physical and chemical characteristics of the host rock and groundwater flow system, and a surrounding geological environment that is likely to remain physically and geochemically stable over long time periods, so that forecasting of long-term behaviour becomes more certain. As discussed above, this is similar to the general type of system that we see in the Cigar Lake analogue that has allowed that uranium ore body to remain in place for over one billion years.

In the case of long-term storage on or near surface, it is likely that the environment surrounding the spent fuel containment system will not be as geochemically or physically stable or unchanging as that found deep underground in a geological repository. Nevertheless, analogues exist that may also give insights in this instance at least for periods of hundreds to perhaps thousands of years. Beyond this time frame, however, natural climatic events such as glaciations, which occur at Canadian latitudes, have destroyed any natural structures that may be considered as surface analogues. The deeper a structure is located underground and the harder the rock it is located in the more resistant it will be to damage from glaciations. Again, analogues such as that found at Cigar lake which have remained intact throughout many glacial periods can provide insights into that ability of systems located several hundred meters underground to withstand glacial events.

4.1 Metals

Metal may be used as barriers in canisters or containers and as reinforcement in concrete. Metals used may be predominantly steels (alloys of iron), but copper and titanium have also been considered in some designs. Several analogue studies have been completed on metals and metal corrosion. These have focused on natural metals such as copper that are thermodynamically stable in certain (reducing or lacking oxygen) geological environments, metals that have been smelted (converted from an oxidized or reduced state to a metallic state) by man hundreds or thousands of years ago and even natural materials, normally thought of as quite reactive or biodegradable, such as wood that if “placed” in a suitable geological environment can be stable over many millions of years. Many barrier material analogue studies have focused on archaeological artifacts rather than naturally occurring metals or materials. There is however a potential bias associated with the study of corrosion of archaeological artifacts, because only the best-preserved specimens survive or are selected by museums for study and it is often not certain what the “weathering” history of these materials has been.

Many common metals such as iron, titanium and aluminum are geochemically very reactive and are seldom if ever found in nature in the metallic state. Other metals such as copper and silver are somewhat less reactive and are often found in the metallic state, as they are thermodynamically stable under certain conditions. Still other metals such as gold and platinum, known as noble metals, are very stable and are commonly found in a metallic state in nature. By understanding the reactivity and thermodynamic properties of metals (as well as economic constraints) from both basic principles and through the use of natural analogues, their properties can be incorporated into the design of a spent fuel management system. The following is a brief discussion of the various types of metals that can be used as barrier systems.

4.1.1 Noble Metals

From a strictly geochemical stability standpoint, “gold plating” of spent fuel canisters for long-term storage or disposal would provide a very chemically stable metallic barrier and there are many natural analogues to demonstrate that gold is chemically stable across a wide range of geochemical conditions. Unfortunately gold is rare and expensive and its use in a repository would potentially make the repository a target for intruders. There are a large number of analogues where gold has been incorporated into structures in the past and its presence has caused these structures to be destroyed or severely degraded due to the actions of those wishing to recover the gold for monetary gain.

4.1.2 Metals of Intermediate Reactivity

Copper is one of the relatively few metals that occurs in its metallic state in the earth. Sometimes, copper nuggets of significant size containing more than 99% copper are found. The largest known deposit like this containing metallic copper is in the Keweenaw Peninsula of Michigan (Crissman and Jacobs, 1982). Here, nuggets of almost pure copper are found in glacial outwash, with only thin protective oxide layers on the exterior surfaces. Also, many bronze artifacts have been recovered from archeological excavations, and these tend to be very well preserved (bronze is an alloy of copper and tin). Data from these and other natural and archeological analogues provide information on copper corrosion rates in both reducing and oxidizing environments, which can then be useful in assessing the potential longevity of metallic copper cladding that may be used in a geologic repository. Several countries including Canada have or are currently considering copper as a protective metal in deep geological repository designs

4.1.3 Reactive Metals

Iron is a good example of a reactive metal. It has been smelted and used as early as 1900 BC by Egyptians (Miller *et al.*, 2000). Perhaps the most comprehensive study on iron corrosion rates under a wide range of environmental conditions was completed by Johnson and Francis (1980). These authors found that the artifacts they studied all tended to corrode at 0.1 to 10 microns annually.

The Inchtuthil Roman nails found in Scotland provide an interesting analogue. At this site, a Roman fortress was abandoned in 87 AD (Angus *et al.*, 1962; Pitts and St. Joseph, 1985). Before retreating to the south, the Romans buried over 1 million nails in a 5-m deep pit, and covered them with 3 m of earth, so the local tribes would not find them. When the nails were unearthed in the 1950s, the nails on the outside of the mass were badly corroded, forming a solid crust of iron oxides (rust) around the remaining mass of nails. The outside layer of nails thus formed a sacrificial redox sink consuming oxygen before it could penetrate to the interior of the mass of nails The physical expansion of the rust as opposed to the original iron also served to self seal the remaining nails from intruding groundwater and oxygen it carried. As a consequence, those nails inside the rusty barrier had minimal corrosion. The survival of the interior nails was thus due to the chemical redox barrier provided by the outer nails, which removed oxygen from infiltrating ground waters before contact with the centre of the mass. In the same manner, large volumes of iron (steel) in waste canisters can be expected to buffer redox conditions in a deep geological repository, removing oxygen carried by infiltrating groundwater, which could otherwise allow oxidizing conditions to develop within the spent fuel. If oxidizing conditions were to develop this may promote the slow dissolution of uranium and other radionuclides and trace elements contained within the spent fuel matrix.

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It is interesting that the same redox processes appear to have occurred in the clays surrounding the Cigar Lake ore body. In this case the outer clay in contact with the groundwater migrating past the ore body is a light reddish colour indicating that the trace amounts of iron in the clay have oxidized. Deeper within the clay the reddish colour disappears suggesting that the oxygen has been consumed and reducing conditions maintained resulting in a very stable ore body.

There are no good natural analogues for other reactive metals that may be used in a repository such as aluminum or titanium, as man has only produced these elements in metallic form within the last couple of hundred years and because of their reactivity they do not exist in metallic form in nature.

4.2 Clays

Bentonite is a group of naturally occurring clays. Bentonite swells when wet and displays high plasticity, has a high chemical sorption capacity (ability to reversibly chemically bind material to its crystalline surface) for most materials and a high thermal conductivity. These properties are why most underground waste repository designs incorporate bentonite as a buffer to surround canisters of spent fuel. Clays do not have a high sorption capacity for some gaseous elements such as iodine and chlorine, which must be immobilized using other materials. When a repository is sealed and groundwater allowed to infiltrate, the bentonite will swell and fill void spaces within the disposal vault. Radioactive contaminants will be able to move through the bentonite by diffusion only, thereby severely restricting their movement from the vault. Bentonite clays also have a high adsorption capacity for many elements and can significantly retard the rate of movement of most elements through them. Clay materials thus can act as very robust physical and chemical barrier systems. This was illustrated above in the discussion of the Cigar lake ore body where the clays formed naturally around the ore body following the deposition of the uranium and acted as a protective barrier for geological time periods.

The Dunarobba Forest in Italy provides another good example of the effectiveness of clays in immobilizing groundwater movement (Benvegnú *et al.*, 1988; Ambrosetti *et al.*, 1992). This forest consists of the preserved stumps of 2 million-year-old trees. The wood has been preserved by the clay surrounding it. The clay prevented oxygenated water from reaching the wood and maintained deoxygenated conditions around the wood. Thus, this clay preserved the wood by preventing groundwater movement around the wood and by maintaining appropriate geochemical conditions within the wood such that bacterial or fungal decay or chemical oxidation could not occur.

In Canada, similar analogues have been found. During the recent mining of diamondiferous kimberlite deposits in the Northwest Territories near the Arctic circle and well above the current tree-line, many pieces of well-preserved wood have been recovered from the kimberlite ore and from shale (consolidated clay) deposits in the same region. When the kimberlite pipes burst through the surface of the earth as a volcanic ash some 50 million years ago fragments of logs fell into the ash and became encased in the ash as it lithified into kimberlite rock over millions of years.

The ash material and rock it became, provided an effective barrier to the infiltration of oxygen and preserved the wood such that not only is its cellular structure intact, but its molecular structure is also largely preserved, making it possible to extract and identify DNA from this 50 million year old material. Studies on this analogue are just underway. In a similar manner, biological materials become buried in clays, which over time consolidate into shales and preserve materials such as wood for geological time periods

5.0 ANALOGUES FOR TRANSPORT AND RETARDATION

Natural analogue studies relating to transport of radioactive elements from a geological repository and through the surrounding geological material have largely focussed on physical and chemical aspects, such as dissolution, diffusion, radiolysis, redox, interaction with colloids, microbial processes, and gas generation and migration. There have been many natural analogue studies relating to transport and retardation, and which cover many specific physical-chemical processes of interest. The reader is referred to Miller *et al.* (2000) for an extensive review. Here, a couple of interesting illustrations of some of these processes are provided.

5.1 Tono Orebody

A deep underground repository in Canada might potentially be located in plutonic rock, which is geologically stable and relatively unfractured. Nonetheless, water will move through rock matrices, and fractures will be present, providing potential pathways for transport of radionuclides, which might migrate from the immediate environment of the repository.

The Tono uranium orebody, located near Tokyo, has been the subject of analogue studies relating to transport of uranium (Miller *et al.*, 2000). The orebody is some 3.4 km long, but was split by a fault some 5 to 10 million years ago. This displaced a portion of the orebody some 30 m upward. The area in general is tectonically active. Despite this large fault, the presence of many other nearby faults and the occurrence of frequent tremors, no significant uranium transport has occurred from the orebody to the adjacent environment. A preferred location for an underground repository would be tectonically inactive, providing even greater physical stability and security than seen at this site.

5.2 Loch Lomond

Loch Lomond, Scotland, provides an analogue, which illustrates the movement of iodine through clays and unconsolidated sediments. Radioactive iodine, especially iodine-129, has a long half-life and its movement from a repository is of interest from a repository design perspective.

Loch Lomond is a land-locked loch (lake) which was flooded by seawater some 6,900 to 5,400 years ago, resulting in a 1-m thick band of marine clay which is now surrounded below and above by freshwater sediments (Falck and Hooker, 1990; Hooker *et al.*, 1985). The concentration of iodine in marine sediments is higher than in freshwater sediments. The concentrations of iodine diminish in the freshwater sediments with distance above and below the marine clay layer. This study provides useful data on diffusion rates for iodine in loosely compacted clay, which can be used to help give confidence in performance assessment of spent fuel repositories, and serves to illustrate that, even after thousands of years, relatively mobile elements such as iodine are escaping to the overlying water only very slowly. This clearly illustrates that so elements such as iodine and chlorine will slowly leak from a repository and the consequences of this leakage must be considered in any safety case made for a used fuel management system.

6.0 ANALOGUES FOR STRUCTURAL MATERIALS

If spent fuel is stored indefinitely at surface, storage procedures might be similar to those presently used for storage of dry fuel after it has sufficiently cooled and decayed in underwater fuel bays. Secure dry surface storage facilities are in place today at Chalk River Laboratories and at Canadian CANDU stations. Such facilities typically use metals and concrete to contain and shield the spent fuel.

On surface storage facilities differ substantially from deep geologic repositories in that they are in oxidizing environments and may be much more exposed to physical and chemical weathering agents. Also, the need for very long-term institutional care and associated governance systems is greater for a surface facility and the integrity of such facilities under future glaciation events in the Canadian environment could be of concern. Security of surface facilities is also a concern. The most relevant analogues for such structures are likely to be man-made structures.

One good archeological analogue that has been considered in the context of concrete materials employed to contain spent fuel, is the concrete found in Hadrian's Wall, built in Britain by the Romans beginning in the first century AD. The wall was manned until abandoned by the Romans in AD 383 Miller *et al.*, 2000). The wall is an analogue not because it has remained intact as a complete structure, as it was largely destroyed by later societies. Rather, it is of interest from the standpoint of the longevity of the cement used to bond the rocks in the wall (Rayment and Pettifer, 1987; Jull and Lees, 1990) Roman cement was somewhat similar to modern Portland cement in that it contains calcium silicate hydrate (CSH) compounds, which provide good strength and bonding capacity. The chemical and mineralogical characteristics of Roman cements provide some assurances about the longevity of modern cements in both surface storage structures designed to last for long time periods and underground storage or disposal repositories. Again as with any analogue, caution must be used, as the conditions under which the material is expected to perform (temperature, radiation fields etc) are not similar. See Figures 4 and 5.

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FIGURE 4 shows a present day view of Hadrian's wall showing how the overall general structure of the wall has survived for close to two thousand years (from Miller *et al*., 2000, reproduced with permission from Elsevier Science).

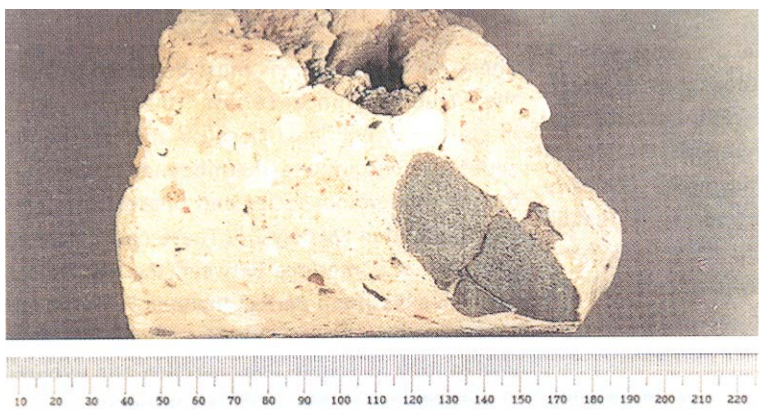

FIGURE 5 shows a close up of some of the cement from Hadrian's wall still retaining its structure and containing encapsulated rocks after almost 2000 years (from Miller *et al*., 2000, reproduced with permission from Elsevier Science)

7.0 SUMMARY

The development of a concept for the long-term management of spent nuclear fuel will of necessity incorporate a number of barriers or containment structures and systems. The degree to which any particular barrier will be relied upon will depend on the particular system and site(s) selected. The particular containment systems range from the composition of the spent fuel itself through to the nature of the surrounding geology in the case of deep geological disposal or alternatively, to the long-term societal governance systems proposed in the event of a long-term storage option.

Many natural analogues have been identified which serve to add to our understanding of materials and processes which will influence the behaviour of radioactive elements in long-term storage or disposal environments. Only a few examples are provided here. Analogues relating to long-term societal governance systems are not discussed in this paper. The analogues discussed take concepts of used fuel disposal from the realm of modelers and scientists to real world examples that can be more broadly understood by members of the general public. Analogue studies are also important for scientists and policy makers charged with developing long-term solutions for used nuclear fuel management, in that they provide insights into and validation for many of the concepts and processes used in their predictive models.

The natural analogues for spent fuel, as best exemplified by the ancient Oklo "reactors", cannot tell us how spent fuel may behave in the short term. They do, however, show that, in the environment that has existed for the past two billion years, uranium and the majority of long-lived fission products and actinides do not tend to migrate away from the site but stay close to their point of origin. The presence of the decay products of shorter-lived (fission-product) isotopes provides indirect evidence that many of these are also not highly mobile. The relative absence of other long lived fission product isotopes such as iodine suggest that these elements can be mobile and must be carefully considered in any safety assessment.

Analogues for decayed spent fuel, which are composed of over 98% by mass of natural uranium can also be exemplified by many uranium ore bodies. In these situations, the analogues have shown that uranium ore bodies remain chemically and physically stable over billions of years in the proper geological environment. The analogues also show that, in other environments, uranium and its decay products can be mobile and move throughout the geosphere and biosphere, both concentrating and diluting as chemical conditions change. By understanding these processes and the factors that drive them we can better site and design spent fuel management systems.

Analogues for how various types of reactive metals such as iron can act as redox buffers and how thermodynamically stable metals such as copper behave under reducing conditions can be valuable. Other barrier analogues from archaeological ruins show that early cements can under the right conditions be stable for thousands of years.

Natural analogues will play a key role in ongoing efforts relating to the design of facilities for used fuel disposal, and in fostering the degree of political and public confidence needed to move forward with the selection of a long-term solution for used fuel management in Canada. Natural analogues do not “prove” the safety of a spent fuel management system. Rather they provide insights into the long-term behaviour of components of a spent fuel management system and can provide a better understanding of their behaviour over long periods of time.

8.0 GLOSSARY

Activation products	Elements that during the course of nearby nuclear disintegrations absorb a subatomic particle and become unstable and radioactive
Analogues	Something that bears an analogy to something else that is similar to it in one or several ways
Becquerel	A measure of the rate of radioactive decay of an element. One bequerel corresponds to one radioactive disintegration per second
Depleted uranium	Uranium as it is mined from the ground in most areas consists principally of two isotopes U-235 (0.73%) and U-238 (99.27%). U-235 is the fissionable isotope that is used to drive the nuclear reaction in power reactors. Light water reactors use enriched uranium in which the U-235 is enriched in the mixture to several percent U-235. The residual U-238 is referred to as depleted uranium as it has been depleted in its U-235 content.
Fissile	Elements such as U-235 split easily when they absorb a neutron and are therefore referred to as fissile or easily split.
Fission	A nuclear reaction in which an atomic nucleus, especially a heavy nucleus such as U-235, splits into fragments, releasing from 100 million to several hundred million electron volts of energy.
Isotopic	One of two or more atoms having the same atomic number but different mass numbers.
Monazite	A reddish-brown phosphate mineral containing rare-earth metals, (Ce, La, Y, Th)PO_4, important as a source of cerium and thorium.
Placer deposits	A glacial or alluvial deposit of sand or gravel containing eroded particles of valuable minerals.
Plutonium	A naturally radioactive, silvery, metallic transuranic element, produced artificially by neutron bombardment of uranium. It's highly fissionable isotope Pu-239, is used as a reactor fuel and in nuclear weapons. Atomic number 94; melting point 640°C; boiling point 3,235°C; specific gravity 19.84; valence 3, 4, 5, 6.

Precipitation	The process of separating a substance from a solution as a solid.
Radionuclides	Nuclides of elements that exhibit radioactivity
Redox barrier	Re(duction) – Ox(idation) or redox refers to reduction and or oxidation chemical reactions that can occur at a particular location. If the redox reaction results in a precipitation event the migration of an element is stopped at the redox barrier or reaction location
Secular equilibrium	When a number of radioactive isotopes in a decay chain are in secular equilibrium it means that they all have the same rate of radioactive decay rate as measured in Bequerels per second.
Sorption	The process of sorbing or bonding in a chemically reversible fashion to a surface.
Spent fuel	Fuel that has reacted in a nuclear reactor and is no longer producing sufficient energy to continue to be economically burned.
Transuranic	Elements that have an atomic number greater than uranium (92)
Uraninite	A complex brownish-black mineral, UO_2, forming the chief ore of uranium and as an ore containing the decay products, protactinium, thorium radium, radium, polonium, bismuth and lead, in secular equilibrium with the parent uranium isotopes. It may also contain traces of other elements.
Zircaloy	A stable, corrosion-resistant zirconium alloy used in the fabrication of nuclear fuel bundles.

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