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ATOMIC ENERGY OF CANADA LIMITED

L'ÉNERGIE ATOMIQUE DU CANADA LIMITÉE

INFLUENCES OF ENGINEERED BARRIER SYSTEMS ON LOW-LEVEL RADIOACTIVE WASTE DISPOSAL

INFLUENCES DES SYSTÈMES DE BARRIÈRE ARTIFICIELLE SUR L'ÉVACUATION DES DÉCHETS RADIOACTIFS DE FAIBLE INTENSITÉ

L.P. BUCKLEY

Presented at the Ninth Annual DOE Low-Level Radioactive Waste Management Conference
Denver, Colorado, August 24-27, 1987

Chalk River Nuclear Laboratories

Laboratoires nucléaires de Chalk River

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RÉSUMÉ

Il y a de grandes différences entre la pratique actuelle de l'ensevelissement à faible profondeur dans le sol et les autres concepts possibles d'évacuation des déchets radioactifs de faible intensité. La protection supplémentaire assurée par les systèmes de barrière artificielle peut apaiser la grande inquiétude du public à l'égard de l'ensevelissement à faible profondeur dans le sol, à savoir: les affaissements de terrain, les eaux souterraines d'infiltration, la migration des radionuclides et la vulnérabilité à l'intrusion des tranchées de faible profondeur.

La présence de diverses barrières artificielles pour réduire l'écoulement des eaux, retenir les radionuclides et empêcher l'intrusion des plantes, des animaux ou des êtres humains conduit à apporter de grandes modifications aux données d'entrée des modèles d'évaluation de l'efficacité. On décrit, dans ce rapport, plusieurs programmes en cours pour la prédiction avec plus de précision de l'efficacité à long terme des barrières artificielles pour déchets radioactifs de faible intensité.

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ABSTRACT

There are major differences between the current practices of shallow land burial and alternative concepts for the disposal of low-level radioactive wastes. Additional protection provided with engineered barrier systems can overcome major concerns the public has with shallow land burial: subsidence; percolating ground waters; radionuclide migration; and, the vulnerability of shallow trenches to intrusion.

The presence of a variety of engineered barriers to restrict water movement, retain radionuclides and to prevent plant animal or human intrusion leads to significant changes to input data for performance assessment models. Several programs which are underway to more accurately predict the long-term performance of engineered barriers for low-level waste will be described.

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INTRODUCTION

The disposal of low-level radioactive waste has not been practiced in Canada. Plans are now underway to begin the disposal of wastes which have been stored and are currently being generated at Atomic Energy of Canada Limited research facilities (1,2). The disposal program has focused on developing technology that will effectively limit water ingress which is the major pathway for radionuclide releases from a disposal facility. Along with site characterization (3), experimental programs are underway to consider radionuclide migration in unsaturated flow regimes in the expected closed system (4).

Disposal of waste at the Chalk River Nuclear Laboratories site will take place in an Intrusion Resistant Underground Structure which has been labelled IRUS. The below ground disposal facility will be located in a free-draining stabilized sand dune above the water table. The facility will utilize four major engineered barriers as a system to prevent water ingress, to restrict radionuclide movement and to reduce intrusion.

The engineered barriers include:

- prepared waste forms
- storage, transport and disposal containers
- backfill and buffer materials, and
- a monolithic concrete cover.

The structure will be designed with 60 cm thick reinforced-concrete walls, and a 100 cm thick self-supporting, reinforced-concrete cover. Backfill will be placed in and around the waste as the waste is positioned within the repository. The unique feature of the facility is the open bottom concept adopted to allow any water which might enter the facility to drain freely rather than remain in contact with the waste. The open bottom will be composed of layers of crushed gravel, compacted sand, clay or other buffer materials. The layers will allow water to pass freely through the repository while retaining radionuclides released from the waste. The various features of the facility are shown in Figure 1.

The concerns most often encountered when discussions about the safety of low-level radioactive waste disposal include the following:

- subsidence of the disposal trench cover;
- percolating water passing through or collecting in the disposal trench;
- subsequent leaching and migration of radionuclides; and,
- the vulnerability of the disposal facility to intrusion.

The concerns raised about shallow land burial and their influence on the research and development programs at the Chalk River Nuclear Laboratories on engineered barrier systems will be discussed.

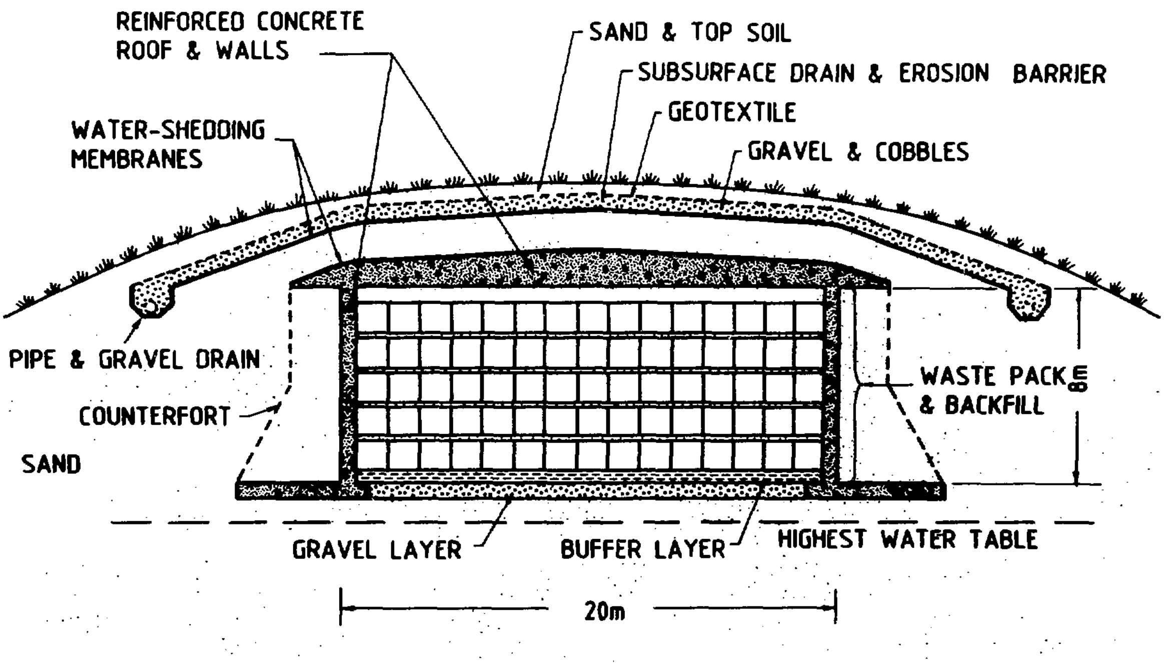


Figure 1. Cross-section of the Completed Intrusion-Resistant Underground Structure (IRUS) for the Disposal of Low-Level Radioactive Wastes.

INTRUSION

The process of disposal of waste requires that some action be taken to restrict or deny entry into the repository once the filling operation has ceased and the facility has been closed. To alleviate plant and animal intrusion, there are a number of barriers which might be employed. One possibility is depicted in Figure 1. In the cross-section of the closed IRUS facility, the use of multiple layers of sand, clay, gravel and cobbles will restrict root travel in the direction of the repository cover and the waste within the repository. Burrowing animals such as rabbits, groundhogs and other smaller species will be thwarted from digging below the zone of gravel and cobbles.

To restrict human intrusion, institutional controls using both active and passive methods will be employed. Fences, groundwater monitoring stations and restricted land use will help to control humans from encroaching on the site. After institutional controls have been removed, the presence of the reinforced concrete cover should deter all but the most determined intruder from entering and dispersing the waste that has been disposed of.

SUBSIDENCE

When the causes of subsidence have been examined, the only experience which can be drawn on is that associated with early attempts to use shallow land trench burial for wastes which were for the most part uncompacted and unconsolidated. Naturally as the waste decomposed, settling of the trench cover took place as the waste volumes contracted. Without support the soil cover collapsed which allowed precipitation to collect in ponds and then percolate into the trench to hasten more decomposition and further collapse of the trench cover. The use of a 100 cm thick reinforced-concrete cover supplemented by water-shedding layers above it is designed to maintain structural stability and prevent ground subsidence even though consolidation of the waste mass will take place. The objective is to ensure that the stability of IRUS is maintained for 500 years or more, and thus avoid subsidence and intrusion until radioactive decay has reduced the inventory of radionuclides such as cesium-137, strontium-90 and tritium to very small values.

The question of durability for a 500-year period will be explored in a study about to commence at the Chalk River Nuclear Laboratories. Degradation of concrete through various mechanisms of external attack from weathering or as the result of internal stresses will be evaluated. The more important and most likely degradation mechanisms will include freeze-thaw cycling, chemical and microbial attack from groundwater or waste decomposition at the surface of the concrete, while degradation from within the concrete could include alkali-aggregate reaction, gel crystallization, differential expansion of the cement and aggregate or by corrosion of the reinforcing steel (5,6). The program will examine these various degradation mechanisms based upon the expected chemical environment, the hydrodynamic conditions expected within and outside the vaults, and the predominant effect of waste decomposition on the chemical degradation of the concrete.

The underground environment at Chalk River in which the repository is to be placed is extremely benign. Once the concrete roof is in place, the structure will be covered to a depth sufficient to keep the concrete from freezing. Below the frost line, the temperature remains between 6 and 10°C year round. The free-draining sand in which the repository is to be placed does not retain much water and dissolution of the cement from the concrete mixture will be an extremely slow process. The groundwater has some carbonate which may weaken the concrete by removal of calcium from the cement paste. Within the repository the low water content expected will inhibit drastic chemistry changes which could alter the integrity of the concrete. Leaching of possible aggressive anions such as sulphates and chlorides from the waste, or production of carbon dioxide from biological processes will be slow. Additives can be chosen and evaluated to make the concrete more impervious to chemical attack. The aggregate chosen for the concrete mixture will be evaluated in the program to ensure alkali-aggregate reactions will not take place, and that there will be no differential expansion between the cement paste and the aggregate to degrade the concrete structure. Good engineering practices in design and construction and a quality assurance program will provide additional assurance once the concrete mixture has been specified to predict a durable lifetime of 500 years.

WATER MOVEMENT

If we first examine how water can enter IRUS, the engineered below ground disposal facility, then one can appreciate how the research and development programs have evolved and are now focused. The repository has been engineered to keep water out. Thus much of the effort is centred on how the repository will behave as designed, that is, unsaturated with water and under reducing conditions. Under these normal conditions some water will be expected from waste decomposition, but otherwise, the repository which is located above the water table will contain only 1-4 % of saturation levels. Under potential failure conditions, such as might be expected from water infiltrating through the cover, or from flooding as a result of a rising water table, parts of the repository may become saturated. When this happens, the water chemistry will become much more complex, and many interactions will take place as depicted in Figure 2. The addition to the water of decomposition products such as fatty acids, the release of chemicals from the solidified waste forms, changes to the buffering capacity of the soil, and backfill materials placed under and around the waste forms, the corrosion of the waste containers, and the release of calcium-based compounds from the concrete will produce yet to be determined effects on the release and migration of radionuclides from the repository to the surrounding environment.

The current studies in understanding water movement in unsaturated and saturated conditions will supplement information to be gained in evaluating the various buffer and backfill materials which will be used to retain radionuclides released from the various wastes to be placed in the repository. Of primary interest is to determine how water will move within the closed repository during unsaturated conditions, how water will be transported as the conditions dictate a move toward saturation, and how the water chemistry will affect the performance of the backfill and buffer

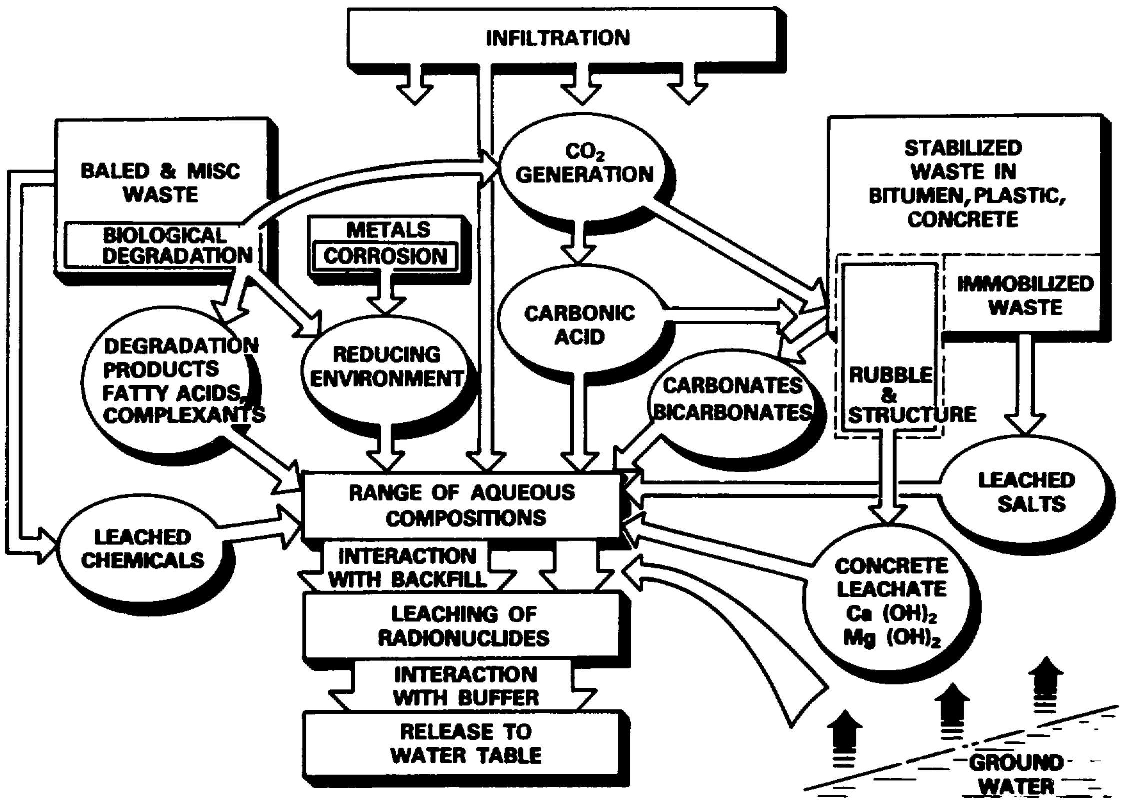


Figure 2. Possible Physical and Chemical Effects on the Repository Environment with Water Uptake and Movement in the Repository.

materials. The determination of the various rate controlling steps is important to establish the effectiveness of the engineered barriers to retain radionuclides and to be able to predict lifetimes of the various components used to retard radionuclide releases.

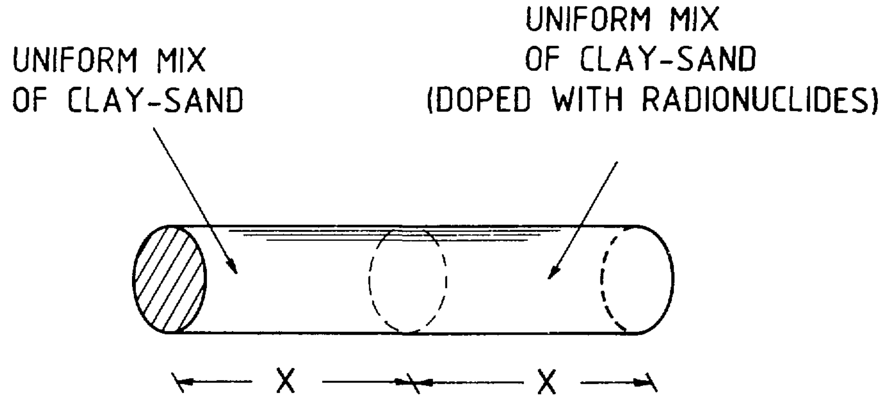
Some of the experiments underway include measurements of diffusion coefficients of water through various combinations of backfill and buffer materials, using gamma scanning techniques on prepared columns of initially dry materials or using pressure plate apparatus for materials containing higher amounts of water. Other experiments involving measurements of ion depletion of the candidate materials is helping to evaluate the rate of deterioration of the materials under different chemistry conditions.

RADIONUCLIDE MIGRATION

The major purpose of putting engineered barriers in place is to restrict the transfer of radionuclides to the environment from the repository. When there is no water flow through a repository, then transport under saturated or unsaturated conditions is dominated by diffusion processes. As the water content diminishes, the diffusion rates should become lower, since the water in the pores between granular particles becomes discontinuous. A number of experiments involving the measurement of the dispersion coefficients of various isotopes (7) indicates such a trend. The experiments are continuing at intermediate levels of saturation and at the much lower levels (1 to 4 % saturation) of unsaturated conditions expected in the closed repository. The experiments are composed of closed cells in which half contains the mixture of buffer material with the water phase doped with radionuclides. A similar mixture without the added radionuclides is placed in contact with the traced mixture (Figure 3). Then over periods of time the cell is counted using non-destructive scanning equipment to yield typical results shown in Figure 4 where normalized concentrations are plotted as a function of axial distance and time. From the expression provided in Figure 3, the dispersion coefficient, D_s can be calculated for a given radionuclide as a function of the water content and the buffer composition.

Experiments, which are being performed in concert with the effect of chemical environment on the aging of buffer and backfill materials, are examining the ability of these candidate materials to adsorb or retain radionuclides. The presence of a high concentration of cations in the vau water could diminish the ability of the materials to selectively retain radionuclides. This could, if the effect were known, be corrected for instance, by increasing the thickness of the buffer material to compensate for lower retention capacity of the buffer material.

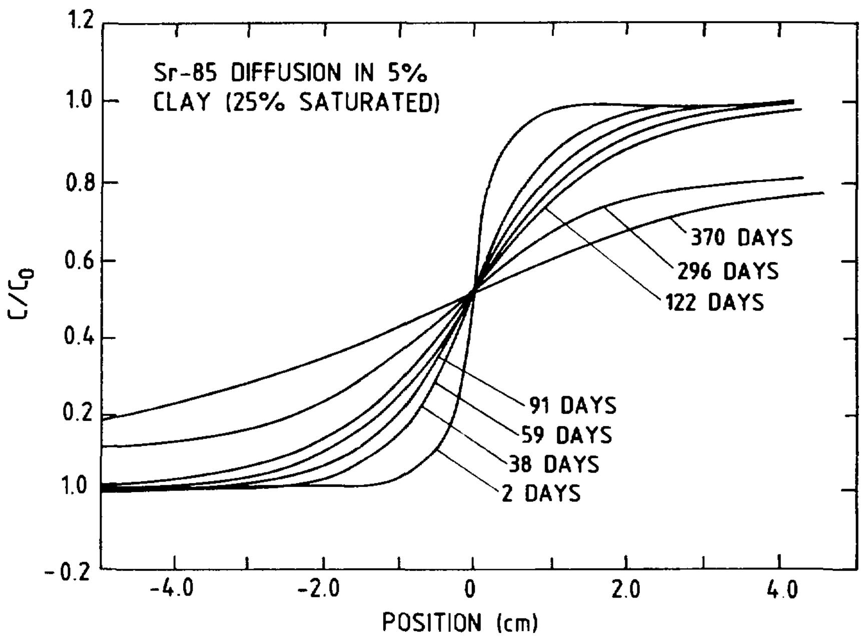
In performing measurements of the leach rates of radionuclides from various matrices, present leach tests such as the ANS 16.1 (8) provide researchers with a tool to compare the performance of various matrices in solidifying a particular waste or to establish optimum processing conditions to yield a consistently high quality product. The leach results from these tests also generate a conservative release rate which can be used in performance assessment models to predict the radiation dose in various intruder scenarios (9). To develop more realistic and thus less conservative release data, several experiments were designed using bench scale lysimeters to



$$\frac{\partial c}{\partial t} = D_s \frac{\partial^2 c}{\partial x^2}$$

$$C/C_0 = 0.5 \text{ erfc } (X\sqrt{4D_s t})$$

Figure 3. Diffusion Experiment for Saturated and Unsaturated Candidate Backfill Materials.



MOVEMENT OF Sr-85 WITH TIME IN A SAND-CLAY MIXTURE

Figure 4. Diffusion of Sr-85 Through a Compacted 5 wt% Clay-Sand Mixture.

evaluate a range of variables representing potential real vault conditions. Among the variables chosen for evaluation were the type of backfill/buffer material, the type of waste form, the influence of different groundwater chemistries and whether or not the lysimeter was saturated or unsaturated. Some of the preliminary results of the test program indicate there is a significant reduction in the release rate from the waste form and when combined with the buffer material the migration of some radionuclides is quite low (10). These studies are continuing, to establish the impact of unsaturated conditions, and the effect of water movement through the lysimeters by infiltration or flooding.

In addition to these bench scale studies, large field scale lysimeters are nearing completion and these will be used for long term verification studies of repository performance in the expected, normal closed environment, as well as the potential failure conditions of infiltration and flooding. As well as attempting to mimic failure modes, the lysimeters will be used to measure tritium movement, to evaluate moisture barriers to be placed over the concrete cover, to use the information gained from observation of radionuclide movement under different water contents, to improve and refine the performance assessment models, and to gain a better understanding of the interaction of all the barriers when they are exposed as a system to changes in environment resulting from waste decomposition, container corrosion, and the addition of chemicals released from the waste.

From sensitivity analyses performed with COSMOS, the performance assessment model developed at AECL (9), long-lived radionuclides such as carbon-14, technetium-99, and iodine-129 in the long term appear to be the major potential contributors to accumulated doses in intruder scenarios. The degree of conservatism in the assessment is difficult to evaluate because of the uncertainties both in the source terms and in the behaviour of these anionic species in disposal environments. Programs are underway to better characterize wastes destined for IRUS, to develop scaling factors for radionuclides, and to investigate the mobility of anionic species within the expected and failure conditions which were outlined above. The interactions of anionic species with barrier materials or selected additives to the buffer zone could greatly assist in lowering the release of these species from the repository and reduce the predicted dose to the inadvertent intruder. Thus some of the experiments described above will be performed with the long-lived radionuclides to establish diffusion coefficients and retardation factors to improve the present input values to the performance assessment code.

SUMMARY

There are major differences between current shallow land burial and proposed engineered disposal systems. Engineered disposal systems offer enhanced protection from intrusion and the virtual elimination of subsidence by the use of a cover of thick reinforced concrete. Engineered disposal systems also will restrict water contact to a much greater extent than is possible with shallow land burial, and by minimizing water movement, radionuclide migration can be greatly diminished. Major research and development programs are underway at the Chalk River Nuclear Laboratories to improve our understanding and to assist the safety assessment analysis of low-level radioactive waste disposal.

We recognize that there is a sparsity of data on concrete durability, and a program is underway to establish the main mechanisms for concrete failure and to determine the rate at which concrete can be expected to fail over a period of at least 500 years when concrete is used as the major barrier to water movement and intrusion from plants, animals and humans.

The present leaching techniques provide conservative estimates for releases of radionuclides from waste forms. Under the expected normal conditions for disposal in engineered facilities, the water pathways will be severely restricted which will result in an unsaturated state. Thus, effort is being spent to establish a release term based on disposal in unsaturated conditions. As well, there is a need to develop an understanding of how radionuclides will migrate in an unsaturated zone, particularly for anionic species which are generally acknowledged to be highly mobile and to use this knowledge in developing better models for performance assessment codes.

The changes with time to wastes placed in a disposal facility, and how the physical and chemical alterations will influence release and migration within the repository are being examined, particularly to evaluate the possible changes in barrier performance as waste decomposes within the repository, containers corrode, and releases of chemicals from concrete and waste takes place.

Finally, while a large amount of effort has taken place in examining barriers on an individual basis, work is now progressing to assess the multiple barriers as a system. The use of both bench- and field-scale lysimeters will attempt to simulate both normal and potential failure conditions of a below ground vault to generate sufficient data that will improve our capabilities to predict future consequences of siting low-level radioactive waste repositories.

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