

## STATUS OF THE CANADIAN NUCLEAR FUEL WASTE MANAGEMENT PROGRAM

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## ABSTRACT

The Canadian Nuclear Fuel Waste Management Program is in the fifth year of a ten-year generic research and development phase. The major objective of this phase of the program is to assess the basic safety and environmental aspects of the concept of isolating immobilized fuel waste by deep underground disposal in plutonic rock.

The major scientific and engineering components of the program, namely immobilization studies, geo-science research, and environmental and safety assessment, are well established.

## INTRODUCTION

Canada's radioactive waste management activities (1) include research programs on highly radioactive nuclear fuel waste (2), low- and intermediate waste (3-6), and uranium mine and mill tailings (7,8).

Research and development pertaining to highly radioactive nuclear fuel waste, which are the subjects of this paper, are performed within the Canadian Nuclear Fuel Waste Management Program. The program covers interim storage, transportation, immobilization, and subsequent disposal of nuclear fuel waste. The term "fuel waste" is taken to mean both used fuel discharged from CANDU\* reactors and radioactive waste that would result from recycling of the used fuel, should recycling be implemented in the future.

In 1978 June, the governments of Canada and Ontario announced an agreement to cooperate in the development of technologies for the safe management and permanent disposal of Canada's nuclear fuel waste. (9)

In 1981 April, the Canadian government approved a ten-year generic research and development program on nuclear fuel waste management. The objectives of this phase of the program are:

- (a) to assess the environmental and safety aspects of the concept of isolating immobilized fuel waste by deep underground disposal in plutonic rock;
- (b) to develop the technology for storage, transportation, immobilization and disposal to the extent necessary to provide data for the assessment; to design facilities; to specify operating processes and procedures; and to demonstrate that practical technology is available for implementation of the concept;

- (c) to establish the requirements, equipment, and procedures for the site characterization and selection processes for the next phase of nuclear fuel waste management; and

- (d) to develop the basis for public acceptance and support through scientific and regulatory review, and public information, interaction and participation.

Under the Canada/Ontario Nuclear Fuel Waste Management agreement (9), responsibility for the development of technologies for interim storage and transportation of used fuel rests with the provincially owned utility, Ontario Hydro, while the coordination and management of the research and development program on immobilization of fuel waste and its safe disposal are the responsibility of the federal Crown Corporation, Atomic Energy of Canada Limited (AECL).

CANDU reactors operate on a natural uranium, once-through fuel cycle. However, technologies are being developed for the immobilization of both used fuel and fuel recycle waste so that either can be disposed of in the future, as requirements demand. Current used-fuel storage methods are adequate for decades, and additional interim storage can readily be provided at the reactor sites. (10) Thus, full-scale disposal is not required in the near future.

To ensure that sufficient technical expertise is available within the program, AECL has actively encouraged the participation of Canada's scientific and engineering community. Several government departments and agencies, private industry and consultants are working with AECL. In addition, faculty members of several Canadian universities have research contracts covering a wide range of topics. Over 400 scientists and engineers are contributing to the program. The administrative structure, main research and development components, participating organizations, and international cooperation are described in more detail in the Program Guide. (11)

Canada has cooperative agreements with the United States of America, the Commission of the European Communities, and Sweden. These agreements provide for the exchange of data and other information on nuclear waste management, and encourage cooperation in areas of mutual interest.

As the halfway point in the ten-year program is approached, it is clear that considerable progress has been made towards achievement of the program objectives. All necessary activities are in place and the remaining steps required to complete this phase of the program have been identified.

\* Canada's natural-uranium-fuelled, heavy-water-moderated and -cooled reactor (CANada Deuterium Uranium).

## ASSESSMENT OF THE CONCEPT

The goal of the environmental and safety assessment is to assess the impact of a disposal facility on man and the environment. The assessments are being published in a series of Concept Assessment Documents. The first interim Concept Assessment Document was published in 1981 (12-14) and the second will be issued shortly. The formal Concept Assessment Document, scheduled for completion in 1988, will form the basis for concept evaluation by regulatory and environmental agencies and for subsequent review at a public hearing.

The environmental and safety assessment has two major components: pre-closure assessment and post-closure assessment. Pre-closure assessment covers the period up to and including vault backfilling, sealing and closure. Post-closure assessment covers the period after the vault has been sealed and the surface facilities decommissioned.

The pre-closure assessment deals with the potential health, environmental and socioeconomic impacts of the activities: construction of a disposal facility; transportation, immobilization and emplacement of the fuel waste; backfilling and sealing of the vault; and decommissioning of the surface facilities. Ontario Hydro performs the pre-closure assessments and has completed documentation of a second interim pre-closure assessment. (15-17) Estimated occupational and public impacts were found to be within currently accepted limits for all operations. Several areas were identified where system modification could further reduce the impacts.

The post-closure assessment considers the potential long-term effects of a disposal vault and its contents on man and the environment after the vault has been sealed. (18,19) Most of the research and development activities focus on the post-closure assessment.

In 1985, February, the Canadian Radiation Protection Association met to consider criteria for nuclear waste management. (20) The meeting included representation from the Atomic Energy Control Board, Atomic Energy of Canada Ltd., and Federal and Provincial Departments of the Environment. Following the conclusion of the meeting two basic criteria for acceptability of a disposal system were adopted, thus facilitating ongoing development of assessment methodology pending the promulgation of formal criteria by the regulatory and environmental agencies. The two criteria are stated as follows:

1. The estimated risk to an individual due to a disposal vault during the post-closure phase should not exceed an established risk level.
2. The estimated probability of exceeding an established individual annual dose equivalent level should not exceed an established probability level.

The above "established" levels have yet to be established, and no conclusion has been reached as to how far in the future the criteria should apply. The first Criterion is considered to be the primary criterion. The Nuclear Energy Agency of the OECD has proposed a risk criterion for nuclear waste disposal and has suggested a level of acceptability of risk at  $10^{-5}$  per annum. (21) Risk is a combination of the probability of receiving a radiation dose and the probability of a health effect arising from that dose.

The second criterion has been adopted to address concerns about high dose levels, even if the first criterion is met. It also enables comparison with natural background dose and current regulatory limits.

Post-closure assessment must draw on vast amounts of research information and data in order to estimate the overall behavior of a disposal vault. This information will concern the behavior of waste forms, containers, buffer and backfill material, the geological formation and the surface environments. In all of these aspects there will be uncertainty and variability-uncertainty because parameters cannot be measured exactly and because their future values cannot be predicted with certainty, and variability because parameter values actually vary considerably in space and time.

The SYVAC systems variability analysis code (Figure 1) was developed to perform the post-closure assessment. (22) SYVAC contains a set of submodels that represent the components of the disposal system, the vault, geosphere and biosphere. Uncertainty and variability are treated by representing the input parameters as distributions rather than single values. The submodels and parameter distributions are derived by assimilating the results of the field and laboratory observations, which are usually interpreted by the use of detailed research models. SYVAC performs repeated deterministic calculations with parameter values sampled from their distributions in a Monte Carlo process. Figure 2 a) and b) contain SYVAC results for a recently completed assessment, comparing risk versus time to the NEA suggested limit and presenting the probability of exceeding 30% natural background dose versus time. (This dose would be equivalent in risk to the NEA risk level.)

Validation of the assessment is achieved by a combination of quality assurance on software (23), expert review, intercode comparison and comparison with field and laboratory observations, the last applying mainly to the validation of the research models. An excellent example of the validation of a research model is the comparison between prediction and observation of the water-table drawdown during construction of the Underground Research Laboratory. (24)

## RESEARCH AND DEVELOPMENT

A comprehensive research and development program is now firmly established (25), which provides the technology required for storage, transportation and immobilization of fuel waste, and for the construction of disposal facilities. It supports the assessments by providing recommended models and data distributions, characterizes waste forms, engineered barriers and natural barriers, and develops procedures and equipment for site characterization and selection.

### Storage and Transportation of Used Fuel

Used CANDU fuel continues to be stored safely and economically in water-filled concrete storage bays at the nuclear generating stations. The current installed nuclear generating capacity in Canada is about 7500 MWe. At the end of 1984, about 300 000 used-fuel bundles (approximately 7 000 Mg) were in storage, after producing about 290 billion kWh of

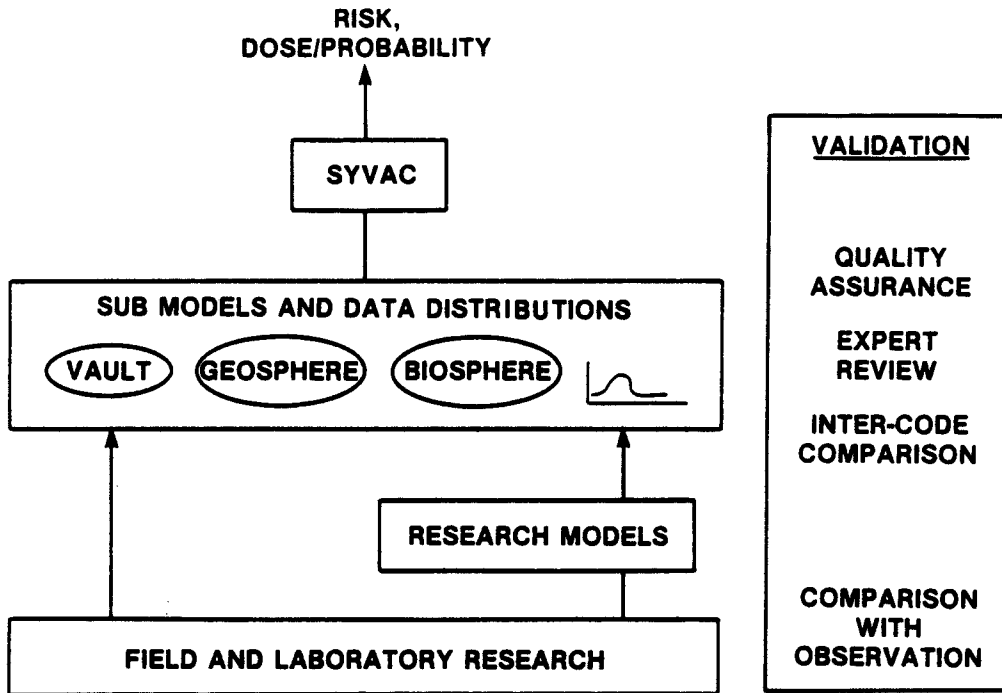


FIGURE 1: POST-CLOSURE ASSESSMENT PROCESS

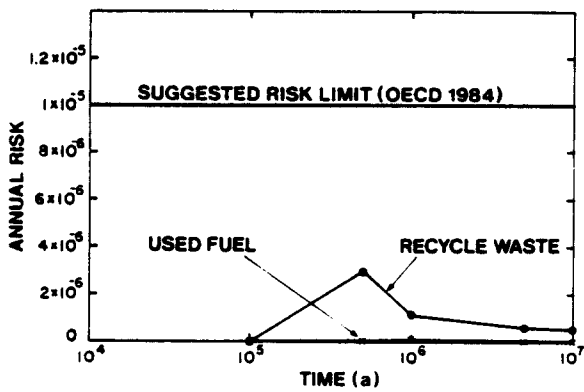


FIGURE 2(a): PREDICTED ANNUAL RISK FROM NUCLEAR FUEL WASTE

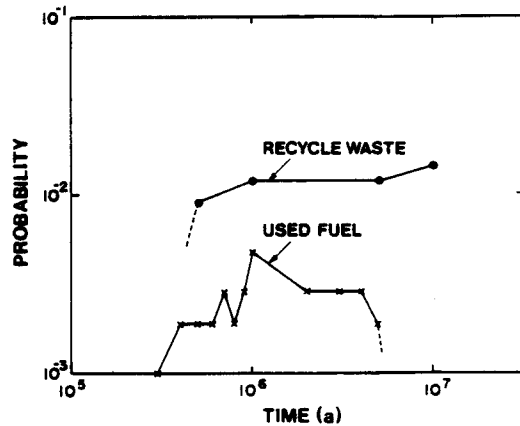


FIGURE 2(b): PROBABILITY OF EXCEEDING 30% OF NATURAL BACKGROUND DOSE

electricity. Studies are being conducted on the feasibility and economics of two dry-storage systems - convection vaults and concrete canisters. Experience with wet and dry storage of used CANDU fuel over the past 20 years provides confidence that interim storage is practicable for at least 50 years. (26)

Ontario Hydro is developing the technology for large-scale transportation of used fuel. The reference cask design has a two-module (192-bundle) payload, rectangular geometry, and monolithic stainless steel wall construction. The heat dissipation capabilities of the reference cask have been investigated in a full-scale simulation using electrically heated simulated fuel bundles, and experiments have been conducted that show that little radioactivity would be released from the fuel in the event of clad rupture during either normal or accident conditions of transport. Completion of design, construction and licensing of a full-size cask is scheduled for 1988.

#### Fuel Immobilization

Fuel immobilization studies involve the development of durable containment for the disposal of intact used-fuel bundles, and the characterization of used fuel as a waste form. (27,28) Studies have concentrated on simple cylindrical containers with a high-integrity corrosion-resistant metallic shell to isolate the fuel during its high toxicity phase. Containment systems that could offer substantially longer isolation, using materials such as ceramics, are also being studied.

Several container designs are being evaluated. (29,30) The simplest, the "stressed-shell" design, has a shell thick enough to withstand the hydrostatic pressure in a flooded vault. Others, called "supported-shell" designs, have an internal support that permits the use of thin-walled shells. The support is provided by a cast metal matrix (for example, lead), packed particulate material surrounding the fuel bundles (for example, glass beads), or a structural support (carbon steel tubes).

Prototypes of these container designs were fabricated from stainless steel or grade-2 titanium and subjected to tests in a Hydrostatic Test Facility (HTF) at pressures up to 10 MPa and temperatures up to 150°C. While short-term tests have shown all containers tested to be acceptable, a detailed structural analysis indicated that a stressed-shell container fabricated from ASTM grade-2 titanium would begin to buckle under creep deformation after about five years under a pressure of 9.4 MPa at 100°C. (31) Modelling of the structural performance of a lead-matrix design has shown good agreement with experiments (32) and voids due to casting defects have been shown not to have significant effects. Hydrostatic testing of full-scale particulate-packed and structurally supported containers has demonstrated excellent resistance to loading, with only minor deformation. (33,34)

Acceptable closure welding has been achieved for grade-2 titanium using tungsten inert gas (TIG) welds (35) and development has progressed on copper container electron-beam welding. (36) Various weld-inspection techniques have been investigated, including ultrasonic. (37,38) Titanium alloys are readily inspectable by ultrasonic techniques but more development work is required for inspecting welds in copper and nickel-based alloys.

The long-term corrosion behaviour of candidate container materials is being studied. (39) Grade-2 titanium has been shown to be susceptible to the initiation of crevice corrosion, but, below a critical potential, the propagation is inhibited. (40) The low levels of dissolved oxygen in deep groundwater, the use of redox buffers and the very limited access of oxidants to the waste containers should all inhibit propagation. It has been suggested that the generation of oxidizing species at the container surface by gamma radiolysis could increase propagation; however, electrochemical experiments have shown little evidence of this so far. (41) It has been concluded that grade-12 titanium is much more resistant to localized corrosion than the grade-2 material (42) as is Hastelloy C-276, a nickel-based alloy.

Hydrogen embrittlement experiments under dynamic-strain conditions have only shown some evidence of embrittlement for Titanium 12 at temperatures above 100°C and at highly oxidizing potentials. (43,44) Studies of the corrosion behaviour of copper in simulated high salinity groundwater have shown that copper is a suitable alternative to passive metals. (45)

#### Used Fuel Characterization

Determination of the leaching and dissolution properties of used  $UO_2$  fuel constitutes the major part of the fuel characterization program. During the past two years, emphasis has been on estimating the fractions of cesium-135 and iodine-129 that are rapidly released from the gap between the fuel and sheath during the early stages of used-fuel dissolution. The gap inventory of cesium-135 and iodine-129 at the time of fuel discharge from the reactors is now estimated to be about 2.2%. Recent studies have shown a correlation between fuel power history and fuel leaching properties. (46)

Experiments to investigate the effects of gamma and alpha radiolysis of groundwater on  $UO_2$  dissolution are underway. After 485 days in a gamma-radiation field at 100°C,  $UO_2$  samples in groundwater became slightly more oxidized than did non irradiated samples. Experiments with alpha sources between 0.037 and 3.7 MBq (1 and 100  $\mu$ Ci) provided no electrochemical evidence for  $UO_2$  oxidation. However, with a 37 MBq (1- mCi) alpha source, electrochemical results suggested that surface oxidation of  $UO_2$  can occur. This is probably due to the reaction of radiolytically produced radicals, or hydrogen peroxide, with the  $UO_2$  surfaces.

#### Waste Immobilization

Processes and products are being developed for immobilizing the waste that would arise if the fuel from CANDU reactors were recycled. (28,47) Glasses, ceramics and glass-ceramics are being evaluated as possible waste forms.

A Waste Immobilization Process Experiment facility, consisting of a rotospray calciner and a ceramic electromelter, designed to produce 10 kg.h<sup>-1</sup> of sodium borosilicate glass, is now operating at WNRE.

The behaviour of glass waste forms and their durability in the hydrothermal environment anticipated in a disposal vault are being studied. A

survey of borosilicate waste glasses (48) showed that durability increases with increasing  $\text{SiO}_2$ ,  $\text{Fe}_2\text{O}_3$  or  $\text{Al}_2\text{O}_3$  content, but decreases with  $\text{Na}_2\text{O}$  or  $\text{K}_2\text{O}$  content. Sodium aluminosilicate glasses (49) have a low, relatively constant leach rate (less than  $10^{-2}$   $\text{kg, m}^{-2} \cdot \text{s}^{-1}$ ) within a wide composition range.

The ceramic waste forms being considered contain sphene ( $\text{CaTiSiO}_5$ ). Calculations indicate that sphene should be stable in groundwaters typical of the Canadian Shield (high  $\text{Ca}^{2+}$ , low  $\text{SO}_4^{2-}$  and  $\text{CO}_3^{2-}$ ) in the temperature range of 25 to  $150^\circ\text{C}$ . Three types of sphene-based matrices are being studied: natural minerals, ceramic pellets formed by pressing and sintering, and glass-ceramics formed by melting and controlled crystallization of the system  $\text{Na}_2\text{O}-\text{Al}_2\text{O}_3-\text{CaO}-\text{TiO}_2-\text{SiO}_2$ . Experimental studies of the hydrothermal behaviour and surface modification of natural and synthetic sphenes in groundwater show that selective leaching of the matrix elements or simulated radionuclides is not significant, provided sphene is within its stability field. (50,51) For the glass-ceramic, consisting of sphene crystallites in an aluminosilicate glass matrix, the release of radionuclides depends on the relative dissolution rates of the glass and ceramic phases and also on the partitioning of each radionuclide between the two phases. (52) Solution and surface analyses indicate that in most cases the glass phase leaches preferentially. For the glass-ceramic, irradiation does not lead to an increase in leach rate in distilled water or brine at  $100^\circ\text{C}$ . (53)

#### Disposal Vault Sealing

Disposal-vault sealing studies involve the development of the buffer material (clay/sand mixture) to surround the containers, and other barriers to close the man-made openings to the surface: namely, the backfill and the plugs and grouts for shaft and borehole seals. (54)

A study of the physical and chemical properties of buffer and backfill clays (55) has provided information on basic mineralogical, chemical and physical properties, and on the behaviour of clays under wet-dry cycling. Bentonites are suitable as buffers because of their high swelling potentials, low hydraulic conductivities, low effective porosities and high sorption capacities for radionuclides. (56-58)

Some important physical properties of candidate buffer materials have now been characterized. A compaction study was completed (59), which showed that the effective clay density in a clay-sand mixture (that is, the ratio of the mass of clay to the volume of clay and voids in the mixture) remains nearly constant for clay contents over 50 weight percent. Effective density is one of the main factors determining the effective porosity, and thus hydraulic conductivity and ionic diffusion properties of the material. (57,60) Swelling pressure has also been shown to be dependent on the effective clay density for one clay-sand mixture. (61)

Hydraulic conductivity values measured for two candidate materials were measured, showed that sodium bentonite clay-sand mixtures have lower conductivity ( $10^{-11}$  to  $10^{-13}$   $\text{m.s}^{-1}$ ) than illite clay-sand mixtures ( $10^{-9}$  to  $10^{-12}$   $\text{m.s}^{-1}$ ). (62) A model was developed to describe the factors (structure, density, water chemistry and hydraulic gradient) that determine the effective porosity of these mixtures. It was

predicted that water chemistry should not significantly affect the porosity for the density values proposed for the buffer. (63)

Mechanical changes can influence the effectiveness of the buffer as a thermal conductor and protective blanket around the waste container. Experimental studies have shown that shrinkage, long-term creep, drying and rewetting, and the removal of buffer material by groundwater are unlikely to prejudice the effectiveness of the buffer. (64-66)

A major study on buffer and backfill engineering has provided information on procedures, schedules and costs. (67,68) The emplacement of containers was assumed to involve four stages: (i) compacting the buffer, (ii) drilling holes into the buffer, (iii) emplacing containers and capping the holes, and (iv) backfilling. Most operations involving backfill and buffer could be carried out by conventional equipment in radiation-free conditions. However, emplacement of the container and capping of the holes would require the use of shielded or remotely controlled equipment.

Computer modelling studies have been performed to determine the effects of container and buffer geometry and the quality of the rock wall in the emplacement boreholes, on diffusional transport of radionuclides from failed containers. (69-73)

#### Immobilized Fuel Test Facility

The Immobilized Fuel Test Facility (IFTF) at WNRE provides an environment for a wide range of multi-component experiments (74,75) in radiation fields, under temperatures and pressure conditions that simulate a vault environment. The experiments are designed to test active waste forms and materials proposed for engineered barriers. Preparation of long-term immersion experiments in passive canisters and of multicomponent-systems tests are well underway. The first set of experiments were emplaced late in 1984. A typical set of experiments comprises 18 small titanium pressure vessels, each containing fuel waste, container material, buffer, groundwater and rock, loaded in one of the seven concrete canisters. The experiments are run for six months or more at temperatures up to  $200^\circ\text{C}$  and at pressures up to 8MPa.

#### Geoscience Research

The emphasis of the geoscience research is on the evaluation of large plutonic rock masses in the Canadian Shield as potential host for immobilized nuclear fuel waste. (76-78)

Deep exploratory drilling and detailed surface mapping are being carried out at designated field research areas in the Canadian Shield. The areas at Chalk River and Atikokan, Ontario, and Whiteshell, Manitoba, contain granite rocks, while those at East Bull Lake and Overflow Bay, Ontario, contain gabbros.

The research area near Atikokan has been chosen as the site of a regional Flow System Study to be carried out over the next eight years. The study area is about 20 km x 20 km and includes the Eye-Dashwa Lakes granitic pluton and a large part of the surrounding rock. Geological mapping of the Eye-Dashwa Lakes pluton and surrounding rock has been carried out. Surficial deposits have been mapped, and the interaction between the shallow groundwater

table and the deeper flow systems has been investigated. Surface water chemistry surveys located electrical conductivity anomalies (up to 300  $\mu\text{S}\cdot\text{cm}^{-1}$ ) that could indicate local zones of recharge or discharge. Sonar surveys of several lakes provided information on the water depth and on the lake bottom sediments.

At the East Bull Lake research area, detailed geophysical surveys were made of the gabbro-anorthosite pluton. (79) The results indicated that both airborne and ground geophysical methods have good predictive capabilities. Estimates of the pluton thickness based on gravimetric (79) and aeromagnetic data (80) were in the range 400 to 800 m, and magnetotelluric surveys (81) suggested the presence of a highly conducting layer at a depth of about 800 m. Subsequent drilling showed that the gabbro-anorthosite layer is about 770 m thick. There was also a good correlation between major faults mapped in the field (82-84) and conductors determined by very low frequency-electromagnetic survey (VLF-EM).

Regional groundwater sampling indicated that saline groundwaters occur at depth throughout the Canadian Shield in fractures in plutons not associated with metallic ore mineralization. The origin of these waters is not yet fully understood, and has been variously attributed to fossil sea water, intense rock-water interaction, and leakage from the Paleozoic cover. (85)

#### Underground Research Laboratory

The Whiteshell research area is situated on the Lac du Bonnet batholith, a large granitic body in southeastern Manitoba. This research area is the site of the Underground Research Laboratory (URL), which is being constructed below the water table in a previously undisturbed portion of the batholith. The URL project has been underway since 1979 (86,87), when field studies commenced to identify a suitable study area and location for the laboratory. In 1980, surface and mineral leases for 21 years were obtained on 3.8  $\text{km}^2$  of Manitoba Government crown land, 15 km northeast of the Whiteshell Nuclear Research Establishment.

The objectives of the URL project are to study the correlation between surface and subsurface features, hydrogeological and geochemical systems in plutonic rock, excavation damage in rock, the effect of heat on plutonic rock (including the effect on mass transport), and heat on buffer/backfill/rock interactions.

Comprehensive geological, geophysical and hydrogeological investigations of the URL lease area are being done. Numerous geophysical surveys were performed in boreholes (88,89), and three major subhorizontal fracture zones were identified. A network of instrumented boreholes has been established to provide baseline data on pre-construction hydrogeological conditions and to measure changes caused by the excavation. Groundwater levels are recorded continuously in about 75 groundwater monitoring locations. Predictions of changes in groundwater systems by several independent hydrogeological modelling groups are being compared with the groundwater system perturbations measured during and after excavation.

Surface facilities are now completed and shaft excavation was completed 1985 March. The URL will be ready for operation in 1986. The underground

facilities have a 255-m deep, rectangular access shaft, a ventilation raise, and a test level with several experimental rooms.

#### Geochemistry and Applied Chemistry

The objective of the geochemistry and applied chemistry research is to quantify the chemical and physical interactions that occur between radionuclides and the geological materials lining waterbearing fractures in plutonic rock. These interactions can prevent or retard migration of radionuclides from the deep underground vault to the biosphere. Examinations of the geological records that exist in and along groundwater-bearing fractures in plutonic rock, and of geological analogues to a disposal vault, such as naturally occurring uranium deposits, are being used to assess the behaviour of radionuclides in the geosphere. In two well-defined uranium deposits in northern Saskatchewan, the uranium was observed to have migrated less than 5 m into the clay surrounding the ore body, over the last billion years.

#### REVIEW PROCESS AND SCHEDULE

In 1981 August, the governments of Canada and Ontario issued a statement describing the evaluation process, the roles and responsibilities of the environmental and regulatory agencies, and the involvement of the public. (90) The evaluation process, which will start in 1988, will involve a regulatory and environmental review, a full public hearing and, in 1991, a decision by the two governments on the acceptability of the concept. In the regulatory and environmental review, the Atomic Energy Control Board will act as the lead agency, assisted by the federal Department of the Environment and the Ontario Ministry of the Environment. The public hearing will be held under the auspices of the Canadian government.

Major efforts have been made to involve the public throughout the program, with speaking engagements, discussion group meetings, and invitations to review assessment documentation. A comprehensive public consultation program is now being launched.

An independent Technical Advisory Committee, established in 1979, provides an ongoing scientific review of the program. The membership of the Committee is drawn from candidates nominated by professional societies throughout Canada, thus ensuring its independent status. The Committee advises AECL on the extent and quality of the program, and interprets and evaluates it for the scientific and technical community and the general public. The Committee also makes constructive criticisms, suggestions and recommendations concerning the various components of the program. The Technical Advisory Committee has issued five annual reports available to the public (see for example, reference 91).

In the second half of the ten-year program it now remains to consolidate the site screening and evaluation methodology, the development of the technology for immobilization and disposal, and the scientific basis for the concept assessment, so that the regulatory and environmental authorities, the public and the various levels of government can reach decisions on the acceptability of the concept.

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