AECL-7787

ATOMIC ENERGY OF CANADA LIMITED

L'ENERGIE ATOMIQUE DU CANADA, LIMITEE

THE CANADIAN NUCLEAR FUEL WASTE MANAGEMENT PROGRAM

PROGRAMME CANADIEN DE GESTION DES DECHETS DE COMBUSTIBLE NUCLEAIRE

T. E. Rummery, E. L. J. Rosinger

Invited paper presented at the International Conference on Radioactive Waste Management, Winnipeg, Canada, 1982 September 13-16

Exposé solicité et présenté à la Conférence internationale sur la Gestion des déchets radioactifs tenue à Winnipeg (Canada) du 13 au 16 septembre 1982.

Whiteshell Nuclear Research Establishment

Etablissement de recherches nucléaires de Whiteshell

Pinawa, Manitoba R0E 1L0

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RESUMÉ

Le Programme canadien de gestion des déchets de combustible nucléaire est maintenant bien établi. Le présent rapport fait état de la recherche et des développements technologiques génériques en cours dans le cadre de ce programme dans le but d'évaluer le concept d'immobilisation et de l'évacuation subséquente des déchets de combustible nucléaire en grande profondeur dans une roche plutonique stable du bouclier canadien. On y fait brièvement mention des participants au programme, des ressources financières, du programme et des procédés de révision externe s'y rattachant. On y donne une description plus détaillée des principaux éléments scientifiques et d'ingénierie du programme, à savoir, les études d'immobilisation, la recherche géoscientifique et l'évaluation de l'environnement de la sûreté.

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ABSTRACT

The Canadian Nuclear Fuel Waste Management Program is now well established. This report outlines the generic research and technological development underway in this program to assess the concept of immobilization and subsequent disposal of nuclear fuel waste deep in a stable plutonic rock in the Canadian Shield. The program participants, funding, schedule and associated external review processes are briefly outlined. The major scientific and engineering components of the program, namely, immobilization studies, geoscience research and environmental and safety assessment, are described in more detail.

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1. INTRODUCTION

The safe containment and disposal of radioactive waste have become important issues throughout the world, and many countries are expending considerable research and development effort in these areas. These national programs, supported and linked by wide international exchange of scientific information, represent a systematic approach by the world's nuclear industries to deal safely and responsibly with their wastes.

Canada's radioactive waste management activities include research programs pertaining to highly radioactive nuclear fuel waste [1-16], to low- and intermediate-level waste [17-20] and to mine and mill tailings [21]. This report outlines generic research and technological development underway to assess the concept of immobilization and subsequent disposal of nuclear fuel waste 500 to 1000 m deep in a stable plutonic rock in the Canadian Shield.

2. CANADIAN NUCLEAR REACTORS

The performance of CANDU* reactors has been, and continues to be, outstanding. In 1981, Canadian reactors held seven of the top ten positions among all nuclear reactors in the western world with gross output over 500 MW(e). In overall lifetime performance, Canadian reactors also occupy seven of the top ten positions.

Canada's first nuclear electric generating station started operation in 1962. In 1983, the net installed nuclear electric generating capacity in Canada will reach 7 GW(e) and by 1990 15 GW(e), when 14 additional reactors planned or under construction become fully operational in the provinces of Ontario, Quebec and New Brunswick.

The Canadian nuclear power program is based on the heavy water moderated and cooled, natural uranium fuelled CANDU reactor which operates on a once-through fuel cycle. At 80 per cent capacity factor, CANDU reactors discharge used fuel at the rate of approximately 140 kg U/(MW(e).a).

The total amount of used fuel now in water-filled storage bays at nuclear electric generating stations is about 6.0 Gg. However, the volume occupied by the used fuel is relatively small. In fact, the 50 Gg of used fuel expected to be produced from all the reactors in Canada by the year 2000 could be stored in bays occupying about the volume of four Olympic-sized swimming pools. Since the storage of used fuel in water bays is adequate for tens of years, and additional storage can readily be provided at the reactor sites [22], commercial disposal is not a near-term requirement.

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* CANada Deuterium Uranium

3. PROGRAM OBJECTIVE, PARTICIPANTS AND FUNDING

The objective of the Canadian nuclear fuel waste management program is to ensure that there will be no significant effects on man and the environment from nuclear fuel waste at any time. Therefore, the goal is to develop technologies for the interim storage and transportation of used fuel, the immobilization of fuel waste, and the permanent disposal of immobilized fuel waste. Under an agreement between the governments of Canada and the province of Ontario announced in 1978, the provincially owned utility, Ontario Hydro, has the responsibility for developing technologies for the interim storage and transportation of irradiated fuel, while Atomic Energy of Canada Limited, a federal crown corporation, is responsible for co-ordinating and managing the research and development program for the immobilization and disposal of fuel waste. Technologies are being developed for the immobilization of both intact used fuel and fuel recycle wastes, so that options are maintained for the disposal of either form.

The immobilization and disposal research program is directed from the Whiteshell Nuclear Research Establishment in Manitoba [23]. Additional expertise is obtained through participation of Ontario Hydro, Energy, Mines and Resources Canada, and Environment Canada. Scientists at ten Canadian universities hold research contracts in various scientific and engineering fields, and private consultants are also extensively involved. Approximately 480 professional scientists and engineers are contributing to the program.

Canada has co-operative agreements in nuclear waste management research with the United States, the Commission of European Communities, and Sweden. Canada is also represented on several technical committees and working groups of the International Atomic Energy Agency and the Nuclear Energy Agency of the Organization for Economic Cooperation and Development [23].

For the ten year period 1981-1990, the Canadian government has approved an average yearly funding of approximately $29 million for generic research and development pertaining to immobilization and disposal. Ontario Hydro, in addition to directing and funding the interim fuel storage and transportation programs, has allocated approximately $2 million in 1982 for technical assistance to Atomic Energy of Canada Limited.

4. RESEARCH AND DEVELOPMENT

The Canadian concept is focused on disposal in a stable, plutonic rock in the Canadian Shield. In the generic research and development phase of the program, we are assessing the basic safety and environmental aspects of a system of multiple barriers designed to prevent radionuclides from the waste re-entering the biosphere in amounts that would present unacceptable risks to man and the environment.

The research and development program at Atomic Energy of Canada Limited has three major components:

1. Immobilization of fuel waste, meaning both used fuel and recycle wastes
2. Geoscience research
3. Environmental and safety assessment

4.1 IMMOBILIZATION STUDIES

For the direct disposal of unreprocessed, used fuel in a geologic vault, studies are concentrating on cylindrical containers employing a high integrity, corrosion-resistant metallic shell to isolate the fuel during its high toxicity phase of about 500 years. Additional studies are considering concepts that offer substantially longer isolation by using materials such as ceramics.

Engineering development includes design, fabrication, testing and assessment of these containers. Specifications include the ability to withstand external hydrostatic loading of the shell to 9.8 MPa (a pressure equivalent to that in a 1000-m deep vault flooded with groundwater) and container temperatures to 150°C, suitability for remote fabrication, inspection and handling, and reasonable cost.

The discovery of highly saline groundwaters within some of the plutonic rock of the Canadian Shield suggests that candidate materials for container construction should have high resistance to corrosion. An assessment of the physical metallurgy, weldability and corrosion performance of a wide range of metals and alloys led to the selection of dilute titanium alloys, nickel-base alloys and copper for detailed study [24]. Type 316L austenitic stainless steel has been used to construct some test containers for preliminary engineering studies.

Several container concepts are under study. The simplest, called the "stressed-shell" design, has a shell of sufficient thickness to withstand the hydrostatic pressure in a flooded vault. Conceptual designs were produced for titanium, nickel-base alloys and 316L stainless steel. A full-scale prototype, capable of isolating 72 fuel bundles, was fabricated from 316L stainless steel, and tested under increasing hydrostatic loading to collapse-failure. Data obtained from strain gauges attached to the prototype's surface were in close agreement with the stresses predicted by a pre-test stress-strain analysis [25].

Other container concepts, called "supported-shell" designs, include some form of internal support, permitting the use of thinner-walled containers. The support is provided either by (i) a cast metal matrix or packed particulate material surrounding the fuel bundles, or (ii) some form of structural support.

Candidate metal-matrix materials include lead, zinc and aluminum/silicon alloy. Various amounts of void will be introduced into the metal matrix to simulate potential casting imperfections. In the packed-particulate concept, material is poured in and vibratory compacted. The principal advantages of the packed-particulate concept are processing at room temperature, lower weight, reduced handling stresses and lower cost. After a series of compaction tests, the particulate selected for further evaluation was 1-mm diameter glass beads.

In the structurally supported design, the fuel bundles are sealed into a rigid inner "basket" of thick-walled carbon steel tubes, which is then inserted into the prefabricated corrosion-resistant shell. Particulate material is used to fill the voids between and around the tubes, and is vibratory compacted.

When all container prototypes have been tested, the performance of the various design concepts will be compared. Eventually, one will be selected as the reference design for more detailed study.

The dissolution and leaching of used fuel are being investigated in detail. Dissolution rates in groundwaters have been studied over a wide range of water chemistry conditions. Under oxidizing conditions at 150°C [26], fuel matrix dissolution is 10 to 20 times greater than at 25°C. The radionuclides Sr-90 and Tc-99 appear to be released by matrix dissolution, whereas Cs-137 is released by a leaching process. Experiments under conditions that are more probable for a disposal vault, i.e. reducing hydrothermal conditions, have begun. Studies of dissolution and leaching of used fuel are being supported by the investigation of the electrochemical oxidation of UO_2 as a function of redox potential and pH [27].

Waste immobilization studies are leading to the development of products and processes for immobilizing, in a durable solid matrix, the radioactive wastes that would arise from recycling used uranium and thorium fuels. A variety of techniques will be required to immobilize the various gaseous, liquid, and solid wastes.

Processes and equipment are being developed for calcination, vitrification and other high-temperature methods for immobilization of non-volatile radionuclides. Methods are also being developed for separating volatile radionuclides from off-gas streams and for their immobilization.

High-level liquid wastes have not yet been generated in large quantities in Canada. Small quantities from the molybdenum-99 production facility at Atomic Energy of Canada's Chalk River Nuclear Laboratories and from the Thorium Fuel Reprocessing Experiment at the Whiteshell Nuclear Research Establishment have been immobilized in borosilicate glass using an in-can melter.

In addition to borosilicates, glass compositions under investigation include aluminosilicates and calcium aluminosilicates (nepheline syenites). Leaching and dissolution processes, including their thermodynamic and kinetic aspects, have been studied under anticipated vault conditions [28,29]. A static leach test in de-ionized water at 100°C has been used to compare the leach resistance of the various glasses. In most instances, the use of de-ionized water ensures worst-case leaching conditions.

The leach resistance of a glass is closely related to its composition and structure [30]. For some borosilicate compositions, an increase in leachant pH, due to dissolution of the glass, can produce a dramatic increase in leaching rate [31]. Conversely, the dissolution rate of many aluminosilicate glasses decreases with time until an apparent constant value is reached, possibly due to saturation of some species in solution.

When granite groundwaters or highly saline groundwaters are used as leachants, some aluminosilicate glasses gain weight due to the formation of hydration layers, or the precipitation of ions from the leachant. In general, under disposal conditions, where the water flow rate is expected to be low, the saturation concentration in the available water may be more important than the leach rate in determining the release of radionuclides to the geosphere.

The now world-famous field test at the Chalk River Nuclear Laboratories, which has been on-going for 21 years and involves 25 lime-nepheline syenite glass hemispheres buried in sandy soil beneath the water table, continues to provide useful data for testing leaching models. The plume distributions of leached strontium-90 and cesium-137 in the surrounding soil were compared with computer predictions using an ion-exchange model [32]. For strontium-90, the predictions and measurements were in good agreement, but for cesium-137, the model did not predict the migration accurately. Recent work on the particulate transport of cesium in groundwater [33] suggests that bacterial action may play a role in cesium transport.

The main advantages of durable glasses as high-level waste forms are their ease of fabrication, their ability to accommodate large variations in waste composition, and their low rate of dissolution at moderate temperatures (< 150°C). However, they are metastable, i.e. they cannot reach true equilibrium in natural waters, and they are therefore susceptible to alteration, particularly at elevated temperatures. Crystalline products are more alteration resistant at higher temperatures (200-300°C), and may reach equilibrium under suitable groundwater conditions.

The crystalline minerals perovskite ($CaTiO_3$) and sphene ($CaTiSiO_5$) are of particular interest, since geochemical evidence indicates that they can take a wide range of foreign ions into their lattices [34]. Studies of thermodynamic stability and dissolution kinetics [35,36] of perovskite suggest that it is less stable than sphene under the disposal conditions anticipated in the Canadian Shield. Sphene is stable at high pH and at the high calcium concentrations found in typical silica-containing groundwaters. A program has been initiated to develop sphene-based glass ceramics, comprising sphene crystallites within a residual sodium aluminosilicate glass matrix [37]. This glass ceramic represents a promising compromise between a glass, with its greater ability to incorporate waste elements, and a purely crystalline matrix, with its greater resistance to radiation damage.

Two options are being considered for the treatment of medium-level waste. The first, direct immobilization in a solid, requires minimal pretreatment (neutralization and evaporation only), and permits the use of several alternative waste forms. On the basis of preliminary results, glass appears to be best for immobilizing medium-level wastes, followed by plastic, bitumen and cement [38]. The second option involves decontamination of the medium-level waste solution with ion-exchange materials, followed by immobilization of the latter in ceramics or glass.

Many of the low-level wastes that would arise from a fuel reprocessing operation are similar to those generated during the operation of

CANDU reactors. Low-level wastes are being catalogued to determine if gaps in technology exist, and if new methods of treatment need to be developed.

In a fuel reprocessing plant, gaseous mixtures containing radionuclides such as tritium, carbon-14, krypton-85 and iodine-129 are released to the off-gas waste stream. Tritium can be immobilized as an insoluble metal tritide [39,40]. Solid hydrated lime and barium hydroxide are being studied as sorbents for carbon-14 [41]. For krypton, selective sorption on zeolitic molecular sieves such as silver- and hydrogen-mordenites is being investigated [42,43]. Two methods of removing iodine are under investigation, one using a corona discharge technique [44] and the other a photochemical abatement process [45]. Both methods remove organic iodides or elemental iodine through precipitation of iodine oxides.

Disposal vault sealing research is concerned with the barriers to be placed between the waste container and the rock, and the barriers that close the man-made openings to the surface. The objective is to provide materials and technology for each barrier, namely, buffer, backfill, and plugs and grouts for sealing (Figure 1), and to assess the effectiveness of each. Bentonite clays are among several materials being examined.

Research includes investigation of additives to the buffer to retard the movement of specific radionuclides, and biological effects in buffer materials. Modeling of mass transport in the buffer and backfill is also under study, and a preliminary evaluation of backfill materials has

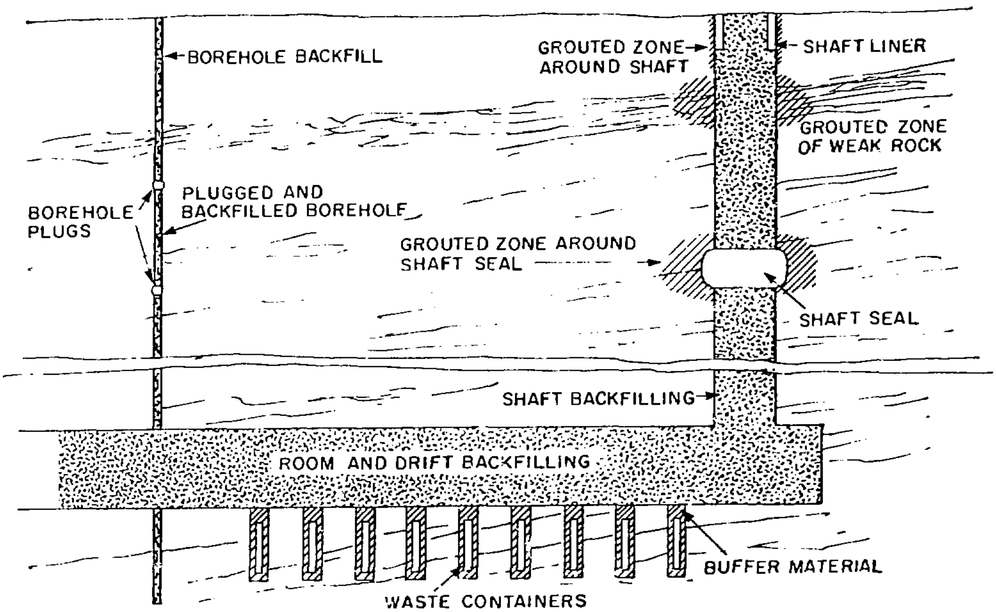

FIGURE 1: Components of Disposal Vault Sealing (not to scale)

begun. Studies of water-uptake and permeability characteristics of backfill, as a function of temperature and composition, are in progress. The applicability of cements and clays for grouting is being investigated. Plugging materials for boreholes and shafts are expected to be similar in composition to those used for the buffer.

4.2 GEOSCIENCE RESEARCH

Geoscience research entails the development of methods for characterizing and quantifying those features of the geosphere that are important in the design and construction of a disposal vault and in the long-term isolation of the waste from man and the environment.

As a result of a recommendation by the Geological Survey of Canada in the early seventies, Canada is directing the main thrust of its geoscience research toward plutonic igneous rocks within the Canadian Shield [46]. A geological map, identifying 1365 plutonic bodies in Ontario that are potentially suitable as disposal sites, has been prepared [47]. However, some effort is being devoted to an examination of alternative geologic formations. As part of the assessment of salt formations in Canada, the Geological Survey of Canada has carried out a literature study of areas in Saskatchewan [48], the Maritimes and Ontario. The suitability of shale as a potential host rock is also being examined.

Field research is underway on rock bodies at five research areas (Figure 2): (1) gneissic rock at the Chalk River Nuclear Laboratories, Ontario, (2) gabbroic rock at East Bull Lake near Massey, Ontario, (3) granite at Forsberg Lake near Atikokan, Ontario, (4) gabbro at Denmark Lake/Overflow Bay near Kenora, Ontario, and (5) granite at the Whiteshell Nuclear Research Establishment and near Lac du Bonnet, Manitoba. Field studies are being performed to quantify major features of the geologic formations, to develop and evaluate equipment and methods for characterizing the formations, and to obtain an understanding of the hydrogeology and geochemistry. Important aspects of the field research are the correlation of results from various test methods and the identification of subsurface characteristics from measurements at the surface and in boreholes.

Our laboratory studies include investigation of the extent to which waste/rock interactions can retard radionuclide migration. For example, we have determined the sorption coefficients of strontium-90, cesium-137, cerium-144, plutonium-237 and americium-241 on core samples from the research areas near Lac du Bonnet and at the Chalk River Nuclear Laboratories. In general, the sorption coefficients were found to increase with increasing mafic content of the rock [49]. On the other hand, the static sorption of strontium by rock samples is reduced substantially in brines. The importance of alteration minerals in the sorption process has been demonstrated using autoradiographic techniques on fracture-fill sections of core from the Forsberg Lake research area [50]. Preliminary observations suggest the following descending order for the degree of sorption: sphene, epidote and chorite, hornblende and biotite, mica, altered plagioclase feldspar, plagioclase feldspar, K-feldspar and quartz. Our

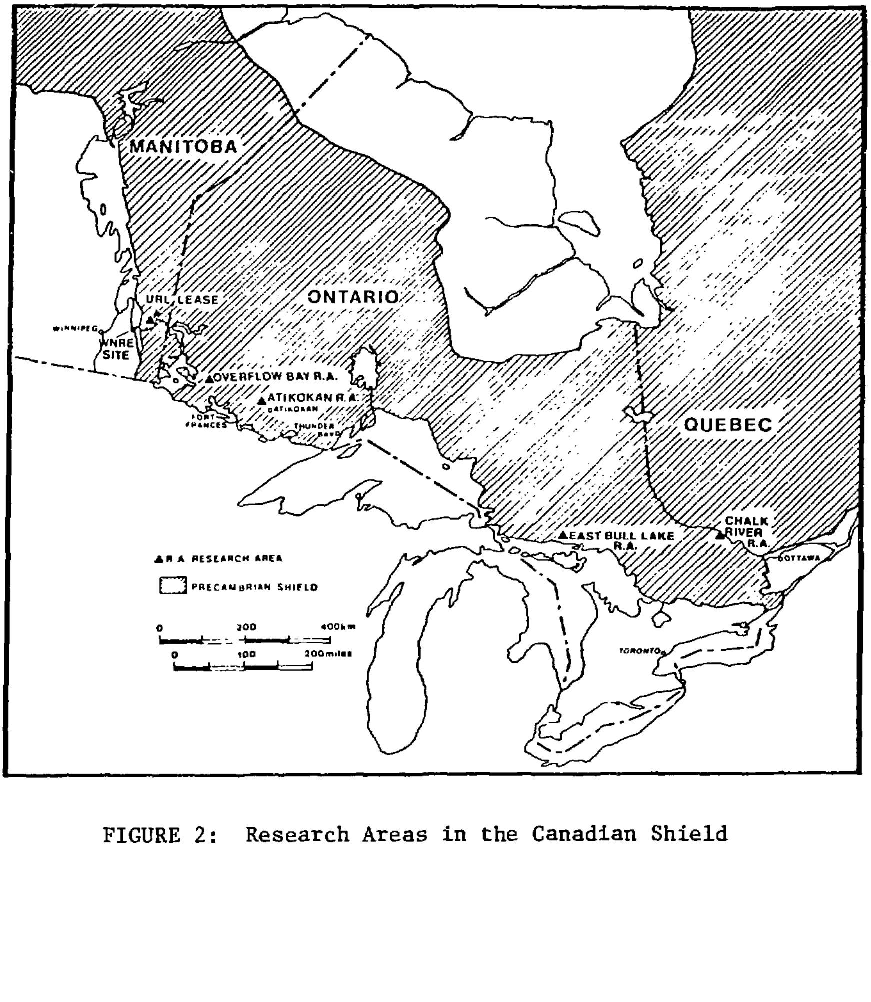

FIGURE 2: Research Areas in the Canadian Shield

results to date suggest that it may be possible to predict radionuclide sorption if the chemical and mineral compositions of the rock are known.

Atomic Energy of Canada Limited plans to construct and operate an Underground Research Laboratory (URL) in the Lac du Bonnet area near the Whiteshell Nuclear Research Establishment (Figure 3). The Laboratory will be the first research facility constructed below the water table in previously undisturbed granitic terrain. After extensive site characterization, laboratory rooms will be constructed at a depth of about 250 m. They will be used for experiments intended to validate predictive models for the performance of a full-scale disposal facility. However no radioactive waste will be used or stored there. A detailed design and contract specifications for the Underground Research Laboratory were recently completed. The

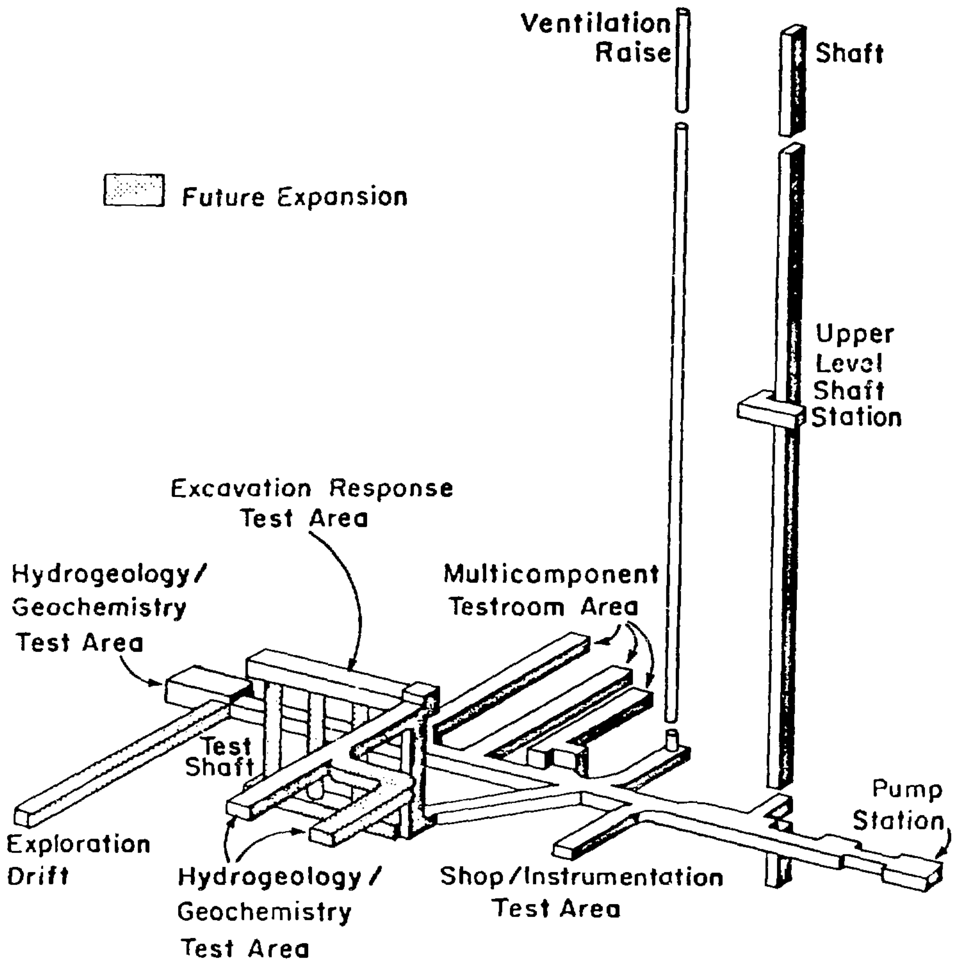

FIGURE 3: URL Underground Layout

surface buildings and services will be build in 1983 and the mine hoist installed. The full operation of the laboratory will start after 1985.

4.3 ENVIRONMENTAL AND SAFETY ASSESSMENT

Environmental and safety assessment involves the estimation of the effects of storage, transportation, immobilization and disposal of nuclear fuel waste on man and the environment. The assessment will form the basis on which endorsement of the disposal concept will be sought from the regulatory authorities, the technical community and the public.

The assessment is divided into two parts, pre- and post-closure [51-53]. Pre-closure assessment considers the impacts of vault construction, transportation of waste, immobilization and disposal operations, until the vault has been backfilled and sealed and the surface facilities removed. It encompasses radiological (pathways) analysis, biophysical/resources analysis, socioeconomic analysis and occupational safety analysis. The post-closure assessment considers the potential long-term effects of the disposal facility on future generations. In the post-closure assessment, the migration of radionuclides from a disposal vault through the geosphere and biosphere, and the effect on man and the environment, are analyzed.

Environmental and safety assessment draws together information from all parts of the program and is based on a systems approach. Laboratory and field research provide data and empirical models, while detailed computer programs help interpret the measured data (Figure 4). The system

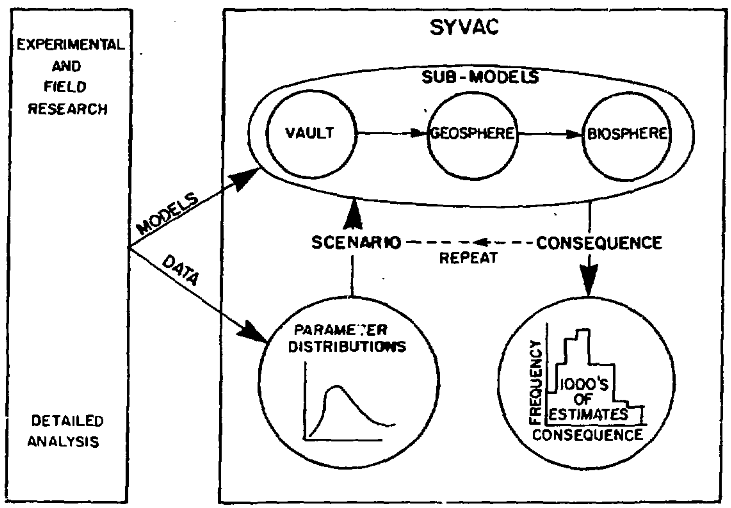

FIGURE 4: Post-Closure Assessment

assessment is performed with the computer program, SYVAC [54], which links a set of sub-models representating the components of the system, i.e. the vault, the geosphere and the biosphere. The sub-models are executed in sequence, leading to an estimate of the consequence for a given scenario as defined below. The consequence is presently taken as the maximum annual radiation dose to an individual in the most exposed group during the first million years after disposal.

Uncertainty in the data and variations in space and time are allowed for by defining the parameter values in the sub-models as distributions, rather than "best estimate" or "conservative" single values. A value for each parameter is sampled from its distribution to form a set. This set of values defines a "scenario". SYVAC then estimates the transport of radionuclides from the vault to the biosphere for this scenario, and determines the consequence (dose to man). Repeated sampling produces different scenarios, for which consequences are determined. The consequences are plotted versus the frequency of their occurrence in the form of a histogram.

The maximum annual radiation dose to man from nuclear fuel waste disposal has been estimated for 1730 scenarios [53]. In 730 cases, the estimates showed no dose in the first million years. The remaining 1000 estimates are shown plotted in Figure 5. For comparison, the natural background dose, one percent of natural background dose, and one percent of the regulatory limit for members of the public for existing nuclear facilities are also shown.

The results from this first assessment are encouraging. In no case did the radiation dose approach that which would be received from

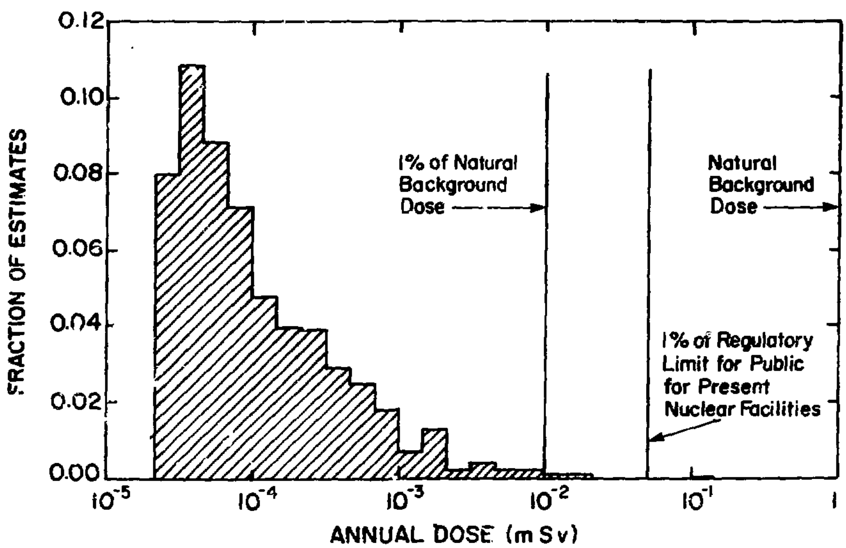

FIGURE 5: Histogram of Maximum Annual Dose

natural background, and in most cases it was much less than 1% of this value. Feedback from reviews, and advances in laboratory and field research are now being incorporated into the next assessment cycle, expected to be completed by September 1983.

5. REVIEW PROCESSES AND SCHEDULE

An informal, ongoing review of the program is obtained by presentation and publication of scientific papers and technical reports. A formal scientific review process is provided by an independent Technical Advisory Committee, the members of which are distinguished Canadian scientists nominated by their respective professional societies. The Committee continuously reviews the research and development program, advises Atomic Energy of Canada Limited on its quality, and makes recommendations for improvement and additional research [56-57].

During the latter stages of the ten-year generic research and development program, an environmental and safety assessment of the disposal concept will be presented to regulatory and environmental bodies for thorough review and evaluation. In August 1981, the federal and Ontario governments issued a statement [58] on the scope of this process, the roles and responsibilities of the environmental and regulatory agencies, and the involvement of the public.

In summary, the evaluation process will involve three stages:

1. Regulatory and environmental review
2. A full public hearing
3. A decision by governments on the acceptability of the concept based on the information and recommendations arising from the first two stages.

The regulatory and environmental review will be conducted by the Interagency Review Committee, which has members from the Atomic Energy Control Board, Environment Canada and the Ontario Ministry of the Environment.

If the federal and Ontario governments approve the concept, the second phase of the nuclear fuel waste management program - site selection for a demonstration disposal facility - can be initiated. The detailed sequence of events and the associated schedule for construction of a demonstration vault and a subsequent commercial facility have yet to be determined.

6. CONCLUSION

The Canadian nuclear fuel waste management program is now firmly established, with a funding level consistent with its technical requirements and a schedule that permits a careful and rigorous development of the concept of disposal deep underground in stable, plutonic rock. Research encompassing all major components of the program is underway, and the review and evaluation process has been developed and defined.

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