PERFORMANCE ASSESSMENT FOR NUCLEAR FUEL WASTE DISPOSAL - THE CANADIAN APPROACH

E.L.J. Rosinger, B.W. Goodwin, D.M. Wuschke
Atomic Energy of Canada Limited
Whiteshell Nuclear Research Establishment
Pinawa, Manitoba ROE 1LO

ABSTRACT

The Canadian concept for nuclear fuel waste disposal involves immobilization of the waste and emplacement in plutonic rock. Predictions of consequences well into the future are carried out with the aid of the SYVAC computer code. SYVAC accepts system descriptions in the form of simplified submodels, and samples input data from distributions that reflect the uncertainty and variability of the data. Current submodels represent the disposal vault, the geosphere and the biosphere. The physical and chemical processes included in each submodel are described. Some results of a performance assessment of the concept are outlined and interpreted.

INTRODUCTION

The Canadian concept for nuclear fuel waste disposal is to immobilize the waste, and to emplace it deep underground in stable plutonic rock[1]. The immobilization technology is being developed for two options: (i) disposal of intact used CANDU fuel and (ii) disposal of the wastes that would result from reprocessing and recycling this fuel.

The assessment of the concept is divided into two major parts: pre-closure assessment and post-closure assessment[2]. The pre-closure assessment considers the period up to and including disposal, and the backfilling, sealing and closure of the disposal facility. This assessment is conventional and will not be discussed in this paper. The post-closure assessment considers the potential long-term effects of the disposal facility on man and the environment after the facility has been closed. This assessment is based on a systems variability analysis approach, developed to perform long-term assessments. The approach is described here, including the models employed, and some recent results are given.

The assessment draws together information from all parts of the Canadian research program; laboratory and field research provide data and empirical models, and detailed computer codes are used to derive simplified system models and to interpret measured data for input to these models.

The post-closure assessment is performed using a computer code SYVAC[3,4,5]. SYVAC contains a set of submodels representing the major components of the disposal system: the vault, the geosphere and the biosphere (see Fig. 1). Parameters that define the behaviour of the system are input as distributions rather than single values. Parameter values are sampled from these distributions to characterize a possible state of the system, i.e. define a "scenario". For each scenario, the transport of radionuclides from the disposal vault to the biosphere is simulated deterministically and a "consequence" is calculated. Repeated sampling of scenarios and calculation of consequences result in a histogram of estimates of consequence versus frequency.

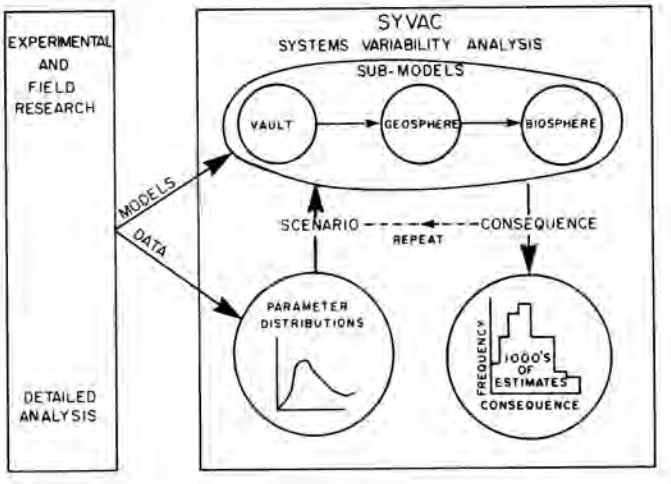

Fig. 1 Schematic representation of the SYVAC computer code and the performance assessment procedure.

An intial version of SYVAC, called SYVAC1, was used in preliminary assessments of used fuel disposal in plutonic rock[6] and of recycle waste disposal under the seabed[7]. Subsequent development resulted in a revised version, SYVAC2, which included extensive modification of the submodels and the executive driver routine[8-11], although the basic concepts of the approach were not changed. New features include consideration of a wide range of waste forms, such as used fuel from CANDU reactors, sodium calcium aluminosilicate and sodium borosilicate glasses, Zircaloy hulls, container infilling materials, and special waste forms for carbon and iodine. The code now contains over 540 sampled parameters and considers the potential detrimental effects of more than 60 radionuclides and chemically toxic elements.

This paper describes the three submodels of SYVAC2, and reports some of the results of the second interim assessment of the Canadian concept for nuclear fuel waste disposal. More details on SYVAC2 and its submodels are available elsewhere[8-11], as are more detailed assessment results[12].

THE SYVAC SUBMODELS

The Vault Submodel

The vault submodel describes the rate of release of radionuclides from the waste forms and waste containers, and their subsequent transport through the buffer and backfill materials to the geosphere[9] (see Fig. 2). The three major processes considered are the release of radionuclides from the waste form, the failure of the waste containers, and the transport of radionuclides within the vault.

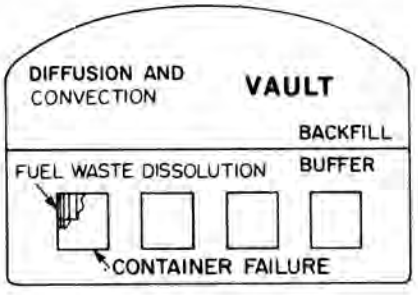

Fig. 2. Vault Submodel

To describe the rate of release of radionuclides from the different waste forms, the vault submodel contains several options. The release of radionuclides from used fuel is described using a two-stage release model[13]. There is a short-term, or "instant", release of iodine, cesium and selenium, because fractions of these elements are present in voids in the used fuel. This release is defined by a parameter called the "instant release fraction", which is described by a truncated lognormal probability distribution with a range of 1.2 to 10%, based on results from a study of the dissolution of used CANDU fuel[14]. For long-term radionuclide release, the solubility of the UO_2 matrix is the controlling parameter. This parameter is described by a truncated lognormal probability distribution[15] with a very wide range, about 10^{-11} to $10^{+0.5}$ mol.m^{-3}.

The release of radionuclides from the sodium calcium aluminosilicate glass waste form is described by a model conceptually similar to that for used fuel. The short-term release is defined by a "leach fraction" parameter, and the long-term release by an "effective glass solubility" parameter[16]. The model for the sodium-borosilicate glass waste form also uses a short-term leach fraction parameter, but the long-term release is defined by a constant leach rate parameter. The release models for the other waste forms are based on the assumption of congruent dissolution, and the use of solubility parameters characteristic of the particular waste form.

To describe the rate of failure of the waste containers, a container failure function (CFF) is used, which gives the fraction of containers failing as a function of time. A detailed study has been carried out to characterize the CFF. It takes into account the spatial and temporal variations of temperature in the waste vault, and uses experimental data for the rates of uniform corrosion of a thin-walled titanium container. The study suggested that the CFF can be described by a truncated normal probability distribution. The uncertainty in the mean lifetime of the container is taken into account by treating it as a parameter selected from a uniform distribution with a mean value of about 1.7×10^4a and a standard deviation of about 3.2×10^3a. The mean lifetime is used to define an early truncation time, before which all containers are assumed to remain intact.

To describe the rate of transport of radionuclides within the vault, a set of time-dependent, one-dimensional equations is used. These equations contain terms for convective and diffusive transport, radioactive decay and equilibrium sorption. Analytical solutions to the transport equations have been derived for three input boundary conditions: a constant concentration, an impulse flux, and a constant flux. These boundary conditions correspond, for example, to congruent dissolution of used fuel, the "instant" release of iodine, and the long-term dissolution of sodium borosilicate glass. The outlet boundary condition at the vault/geosphere interface uses a mass-transfer coefficient that allows coupling of the vault and the geosphere. The solution to this set of equations then corresponds to the transport of radionuclides from one waste container to the geosphere. This output flux is then convoluted with the CFF to give the total radionuclide flux from the vault to the geosphere as a function of time.

The Geosphere Submodel

In the geosphere submodel, the transport of radionuclides through the geosphere is represented by a set of time-dependent, one-dimensional equations containing terms for convective transport, dispersion and molecular diffusion, radioactive decay and equilibrium sorption[10] (see Fig. 3). Dispersion and molecular diffusion are described by a single parameter, the hydrodynamic dispersion coefficient, which is assumed to be constant for a given scenario. An analytical solution to this set of equations has been derived for an impulse flux input and a semi-infinite outlet boundary condition. Convolution of this solution with the output flux from the vault yields a time-dependent output flux from the geosphere.

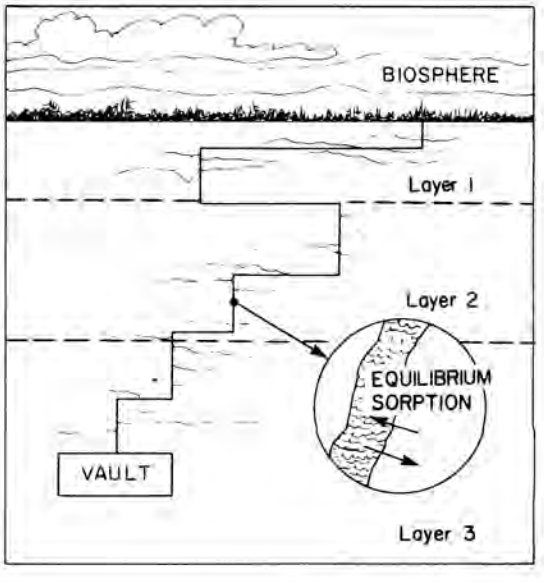

Fig. 3 Geosphere submodel.

Some parameter values for the geosphere submodel are selected from representative values for three separate horizontal layers. (The uppermost layer represents the weathered portion of the plutonic rock, and the lower layers are characteristic of deep plutonic rock). As an example, the hydraulic conductivity is described by different probability distributions for each of the three layers. Parameter values are selected for each layer and are used to estimate a set of "equivalent" parameters for the whole geosphere. The equivalent parameters, such as for groundwater velocity and path length, are then used in the analytical solution describing the rate of transport of radionuclides.

The Biosphere Submodel

The biosphere submodel consists of transport and dose components[11] (see Fig. 4).

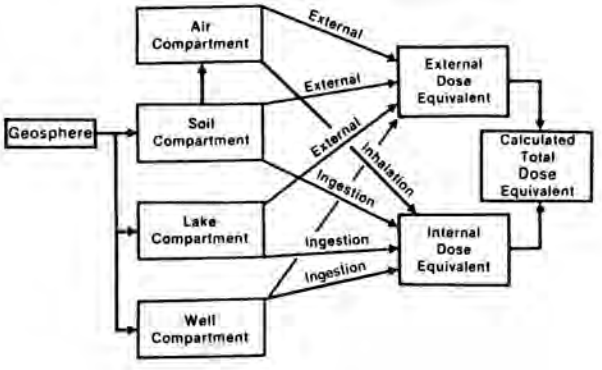

Fig. 4 Biosphere submodel.

The transport component is used to calculate time-dependent concentrations of radionuclides in air, soil, lake and well compartments. Concentrations in the well compartment are calculated from concentrations in the geosphere, modified by a dilution factor related to the volume of water that a typical well must produce to be viable. Concentrations in the air compartment are determined from the time-dependent concentrations in the soil, via an atmospheric dust-loading approach.

The dose component of the biosphere submodel is used to calculate the radiation dose equivalent for ingestion and inhalation of radionuclides, and for external exposure. For food ingestion, a food-chain model based on man's energy requirements is used[17,18]. The source of drinking water is assumed to be a freshwater lake, or a well. The frequency of choosing a well is based on estimates of the use of wells in the Canadian Shield. Inhalation of radon gas and of particulates containing radionuclides is also modelled. For determining external exposures, radionuclide concentrations in the air, soil, well and lake compartments are considered.

The Executive Modules

The three submodels described above are interconnected within the SYVAC2 code by a group of modules called "executive" modules. These modules input the parameter distributions and select the set of parameter values required to define a scenario. The executive modules also perform the convolution integrations that link the output of one submodel to the input of the next (such as the output of the vault to the input of the geosphere), and save results from each deterministic simulation for later analysis and interpretation.

ANALYSIS AND INTERPRETATION OF SYVAC RESULTS

SYVAC2 has been applied to the assessment of the consequences of disposal of used fuel and fuel recycle waste[12]. Here, consequence is defined as "the maximum annual effective dose equivalent to a member of the most exposed group, occurring up to a specified time". "Zero" consequence is defined as an annual dose less than 10^{-10} mSv, a level about ten orders of magnitude lower than the dose from natural background radiation. Consequence is hereafter referred to as "maximum annual dose equivalent" or, simply, "maximum dose".

Assessment of the Disposal of Used Fuel

The histograms in Fig. 5 show the distributions of maximum dose for periods up to 10^5, 10^6 and 10^7 years for the disposal of used fuel. The percentage of simulations giving non-zero doses was 3%, 33%, and 92% for 10^5, 10^6 and 10^7 years, respectively. No doses greater than 10^{-10} mSv were received until 24 000 years.

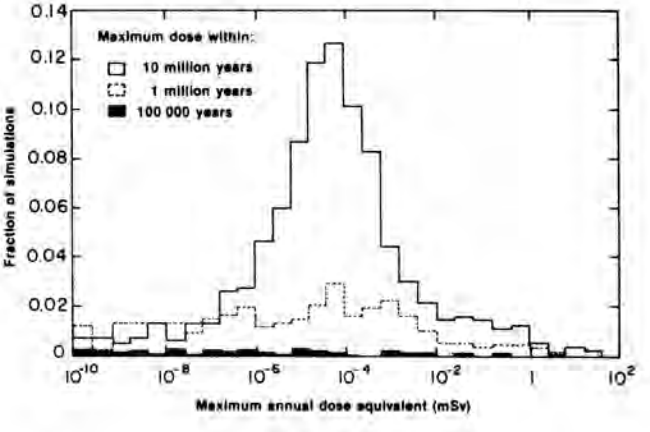

Fig. 5 Distributions of maximum annual dose equivalent for used fuel disposal.

Another method of presenting the results is shown in Fig. 6, where the downward cumulative fraction of simulations is plotted against the maximum annual dose equivalent. The fraction of simulations giving doses exceeding any specified level can be read directly from the plot. For example, the fraction that exceeds 10^{-2} mSv is about 7% for the case of 10^7 years.

In Fig. 7, distributions of total maximum dose from all radionuclides and maximum doses from individual radionuclides are plotted for the 10^7 year case. These plots show that ^{129}I is the only radionuclide producing annual doses greater than 10^{-3} mSv. ^{129}I contributed to maximum dose in all 92% of the simulations that resulted in non-zero doses, while ^{99}Tc, ^{242}Pu and the other radionuclides contributed to maximum doses in 6%, 1% and 0.7% of the simulations, respectively.

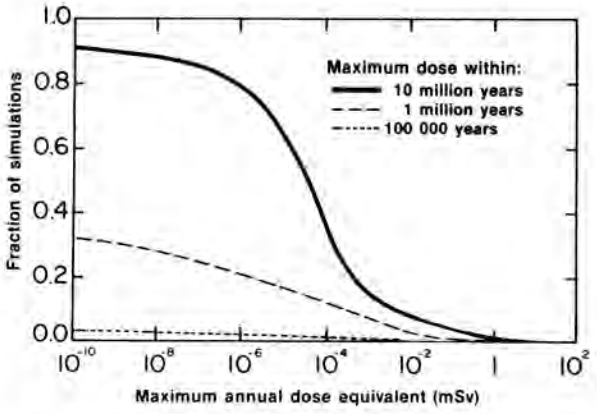

Fig. 6 Downward cumulative distributions of maximum annual dose equivalent for used fuel disposal.

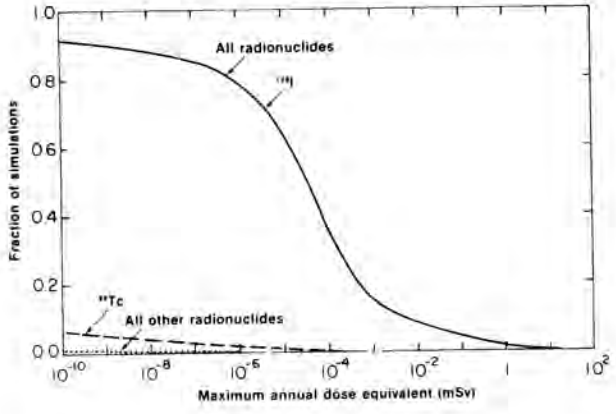

Fig. 7 Downward cumulative distributions of maximum total annual dose equivalent and maximum doses from individual radionuclides within 10^7 years, for used fuel disposal.

The relative contributions of the ingestion, inhalation and external pathways to total dose were compared. The dose distributions for ingestion and total dose were essentially the same, indicating that the ingestion pathway is much more important than the inhalation or external exposure pathways.

Further study of the ingestion pathway included an analysis of the importance of the well-water pathway. In about 10% of the simulations, the source of man's and animals' drinking water was a deep well extending into bedrock. Fig. 8 shows the distribution of maximum dose for all simulations for the 10^7 year case and the distribution of doses when drinking water is obtained from a well or a lake. All simulations giving doses higher than 0.3 mSv involve the well-water pathway.

The low doses arising from most radionuclides and the relatively high doses from ^{129}I can be attributed primarily to the performance of two barriers: the used fuel waste form, which limits the release of radionuclides from the vault, and the geosphere, which delays the transport of radionuclides to the environment.

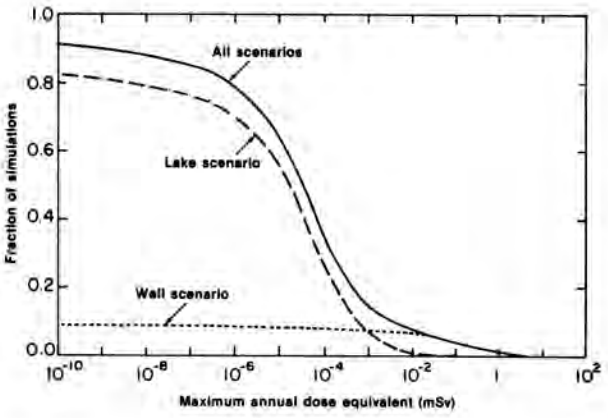

Fig. 8 Downward cumulative distribution of maximum annual dose equivalent within 10^7 years for used fuel disposal, and annual dose equivalent distributions for the lake and well scenarios.

Most radionuclides are uniformly distributed in the UO_2 fuel matrix, and are assumed to be released only by dissolution of this matrix. This is an extremely slow process, as illustrated in Fig. 9, which shows the fractions of radionuclides released by congruent dissolution of the UO_2 matrix within 10^7 years. However, fractions of a few radionuclides, ^{79}Se, ^{135}Cs and ^{129}I, which are present in the voids in the used fuel, are assumed to be released instantly upon container failure. Since most containers fail well before 10^7 years, the fractions of these radionuclides released are essentially equal to those present in the voids. The median value of these instantly released fractions is about 3×10^{-2}, more than seven orders of magnitude greater than the median value for the fraction of radionuclides released by congruent dissolution of the UO_2 matrix. Furthermore, the rate of release of these fractions from the vault is much greater because it is controlled by the rate of container failure, and containers are predicted to fail during a relatively short interval, 2000 - 10 000 years.

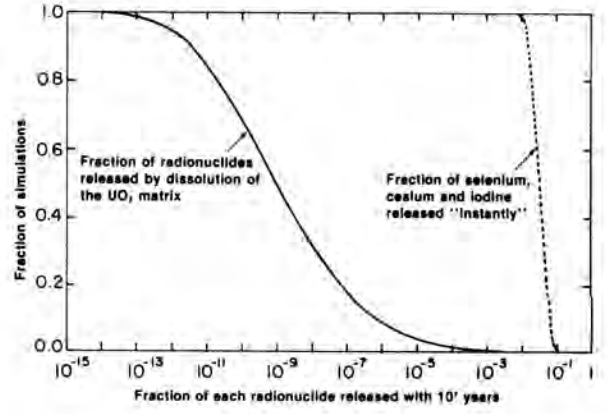

Fig. 9 Fractions of radionuclides released from used fuel within 10^7 years.

The maximum dose received also depends on the length of time required for the transport of radionuclides to the environment. This transport time depends on the time required for groundwater to travel through the geosphere and on retardation of radionuclides in the geosphere. The groundwater transit time is generally very long, with a median value of 1.2×10^7 years (see Fig. 10). However, the range of values is large, with the minimum being about 70 000 years. Because of hydrodynamic dispersion, radionuclides can travel through the geosphere in less than the groundwater transit time. This accounts for a few low doses being predicted at times as early as 24 000 years.

Additional delay in the arrival of some radionuclides at the biosphere would be provided by interaction of radionuclides with rock-fracture surfaces. Also, many radionuclides would decay while being transported by groundwater, so that only insignificant quantities would reach the biosphere. An exception is ^{129}I, which has a very long half-life, and which is assumed to undergo no interactions with the geosphere. The combination of these properties and its high rate of release from used fuel make ^{129}I the most important contributor to dose.

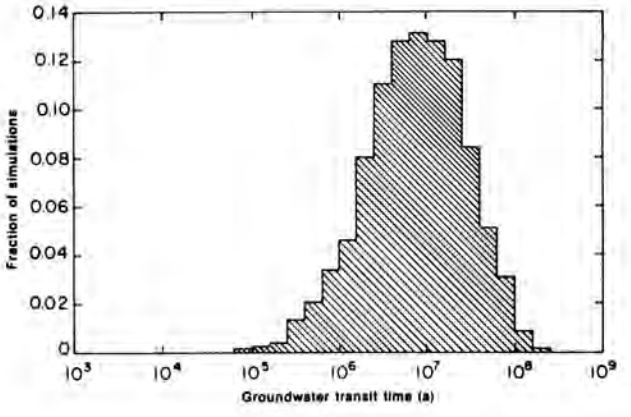

Fig. 10 Distribution of groundwater transit times.

Assessment of Disposal of Fuel Recycle Waste

SYVAC2 has also been used to assess the disposal of fuel recycle waste. This disposal system differs from that for used fuel in the initial inventory of radionuclides, in the waste forms and their ability to retain radionuclides, and in details of the vault geometry. By far the most important difference is the ability of the waste form to retain radionuclides. For the analyses presented here, the fuel recycle waste forms were as follows:

(1) sodium calcium aluminosilicate glass, in which most of the fission and activation products are incorporated;
(2) bismuth oxyiodide, containing ^{129}I;
(3) barium carbonate, containing ^{14}C, and
(4) compacted Zircaloy fuel-bundle components.

Figure 11 shows the distribution of maximum annual dose equivalent within 10^5, 10^6 and 10^7 years for this combination of fuel recycle waste forms. For the 10^7 year case, the fraction of maximum doses exceeding 10^{-2} mSv is 2.5% and the fraction of non-zero doses is 37%. No doses greater than 10^{-10} mSv were received until 13 000 years. As illustrated in Fig. 12, ^{99}Tc is the most significant radionuclide and the only one producing maximum doses greater than 10^{-3} mSv. ^{129}I is the second most significant radionuclide, while ^{242}Pu and about 25 other radionuclides produced only very small maximum doses. A single unusual simulation, where the groundwater transit time was 136 years, was the only exception. For this simulation, five radionuclides produced maximum doses exceeding 10^{-3} mSv, and one, ^{229}Th, produced a maximum dose of 56 mSv.

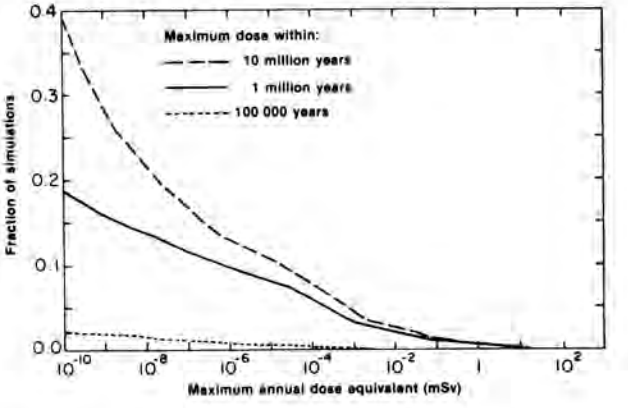

Fig. 11 Downward cumulative distributions of maximum annual dose equivalent for fuel recycle waste disposal.

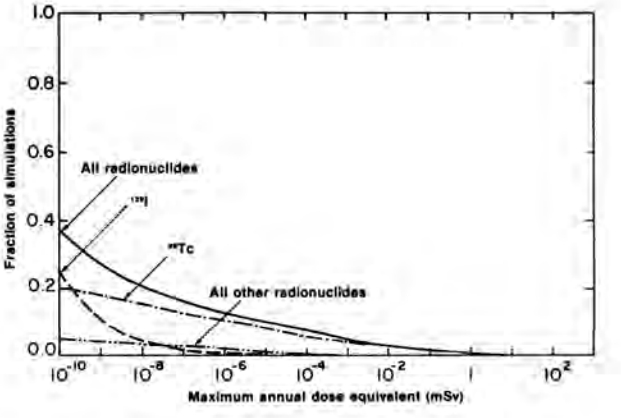

Fig. 12 Downward cumulative distributions of maximum total annual dose equivalent and maximum doses from individual radionuclides within 10^7 years, for fuel recycle waste disposal.

The external and inhalation pathways are insignificant compared to the ingestion pathway. Analysis of the importance of the well-water pathway showed that the maximum dose distribution was nearly independent of the source of drinking water. This was because, for fuel recycle waste ^{99}Tc was the most important radionuclide and, for ^{99}Tc, ingestion of water is unimportant relative to ingestion of terrestrial plants.

The performance of the different recycle waste forms is illustrated in Fig. 13. The median value for the fraction of radionuclides released within 10^7 years from the sodium calcium aluminosilicate glass is approximately 10^{-3}, about six orders of magnitude higher than the corresponding value for congruent release from the UO_2 matrix. However, retention of ^{129}I by its recycle waste form, bismuth oxyiodide[19], is much better than that by used fuel. The median value of the fraction of ^{129}I released from bismuth oxyiodide is about 4×10^{-8}, nearly six orders of magnitude lower than the median value for release from the voids in used fuel.

The importance of the durability of the ^{129}I waste form is illustrated by the third curve in Fig. 13, which shows the distribution of the fraction of ^{129}I that would be released from barium iodate, an alternative waste form for ^{129}I. The median value is 0.9, about two orders of magnitude higher than that for release of ^{129}I from used fuel.

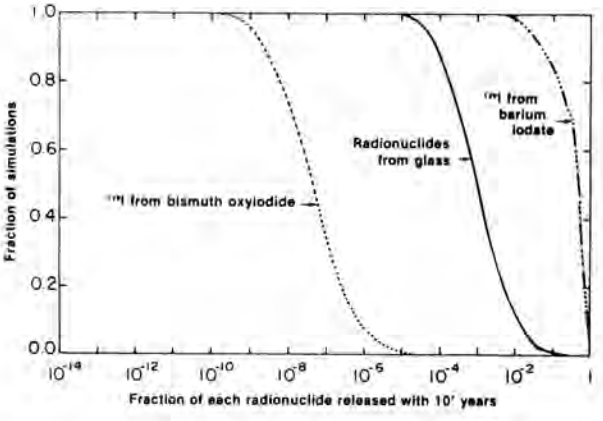

Fig. 13 Fractions of radionuclides released from different fuel recycle waste forms within 10^7 years.

Figure 14 illustrates how the distribution of maximum annual dose equivalent for fuel recycle waste is affected by changing from a bismuth oxyiodide waste form to barium iodate. Maximum doses were generally higher for the barium iodate waste form than those for bismuth oxyiodide. For example, the fraction of doses exceeding 10^{-2} mSv increased from 3% to 11%. Both ^{129}I and ^{99}Tc were important contributors to maximum dose.

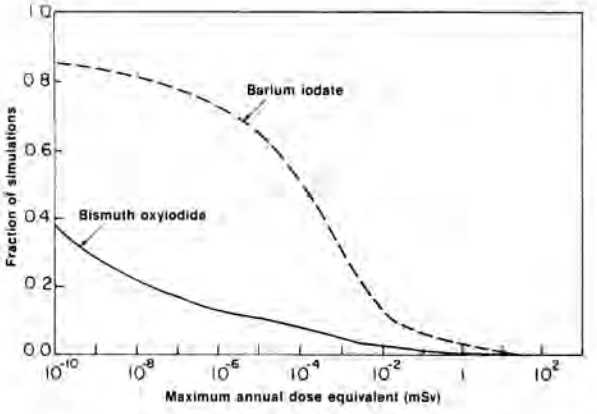

Fig. 14 Downward cumulative distribution of maximum annual dose equivalent within 10^7 years for different fuel recycle waste forms.

SUMMARY

The SYVAC2 submodels described in this paper have undergone substantial changes from the original SYVAC1. These changes are mostly in the types and details of processes modelled, and not in the SYVAC concept itself. The trend for SYVAC3, the next version of the SYVAC code, is to consolidate the submodels, and to ensure that all models and data are soundly supported by results from the R and D programs.

Assessment of the long-term impact of the disposal of nuclear fuel waste using SYVAC provides a great deal of information about the behavior of the disposal system. However, since development of the submodels is not yet complete, and detailed experimental data are not available for some parameters, the SYVAC2 results should be considered as preliminary.

The results have been presented for the disposal of nuclear fuel waste in the form of

- used fuel bundles,
- fuel recycle waste, with ^{129}I incorporated in bismuth oxyiodide waste form, ^{14}C in a barium carbonate waste form, other radionuclides in sodium calcium aluminosilicate glass, and compacted Zircaloy fuel-bundle components,
- fuel recycle waste, with ^{129}I incorporated in a barium iodate waste form, ^{14}C in a barium carbonate waste form, other radionuclides in sodium calcium aluminosilicate glass, and compacted Zircaloy fuel-bundle components.

The assessment yielded the following results:

(1) For all of the above three options, no consequences are predicted for tens of thousands of years after disposal.

(2) For all three options, the highest predicted maximum doses were similar. The fraction of maximum annual dose equivalents exceeding 10^{-2} mSv was 7%, 3% and 11% for used fuel, recycle waste with ^{129}I in bismuth oxyiodide, and recycle waste with ^{129}I in barium iodate, respectively.

(3) For used fuel disposal, ^{129}I was by far the most important contributor to maximum dose, and the only radionuclide producing maximum doses greater than 10^{-3} mSv. For recycle waste with ^{129}I in bismuth oxyiodide, ^{99}Tc produced the highest maximum doses and was also the major contributor to maximum dose, especially in the high dose region. For recycle waste with ^{129}I in barium iodate, both ^{129}I and ^{99}Tc were important.

(4) For all three options, the doses received were almost entirely via the ingestion pathway. For used fuel disposal, the well-water pathway produced the highest doses.

Review and consolidation of SYVAC submodels will be complemented by sensitivity analysis studies and quality assurance of the software. SYVAC3 is under development and is scheduled for completion later this year. The third interim assessment of the Canadian disposal concept will be presented in 1986. A formal concept assessment document, to be issued in 1988, will provide a focus for formal evaluation of the disposal concept via regulatory review and a public hearing.

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