Estimating Gas-Borne Dose Consequences from a Used Fuel Repository in Sedimentary Rock

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The Nuclear Waste Management Organization (NWMO) is responsible for the implementation of Adaptive Phased Management (APM), the federally-approved plan for safe long-term management of Canada's used nuclear fuel. Under the APM plan, used nuclear fuel will ultimately be placed within a deep geological repository in a suitable rock formation.

Before specific sites have been identified for detailed examination, generic studies are used to illustrate the long-term performance and safety of the multi-barrier repository system within various geological settings. The NWMO recently completed a study illustrating the postclosure safety of a conceptual deep geological repository located in the sedimentary rock of the Michigan Basin in Southern Ontario.

Exposure of the steel components of an underground repository to groundwater can result in the generation of hydrogen gas due to corrosion processes. The fate of this gas, as it migrates through the repository, the excavation-damaged zone (EDZ), and the surrounding host rock can affect both the internal repository pressure and the transport of gaseous radionuclides. The results presented here focus on gas generation modelling, transport of volatile radionuclides, and the gas-borne dose consequences from C-14.

A conservative assessment of gas generation is determined using a hypothetical disruptive scenario in which all containers fail after 10,000 years. In this case, the steel in all containers is exposed and starts to corrode. A bounding estimate of the dose consequence due to C-14 is obtained by assuming the gas is not dispersed, that all C-14 exits the repository via a single shaft, and that all C-14 enters a house that is built directly on top of the shaft. Based on these extremely conservative assumptions, results show that gas-borne dose consequences will not exceed the interim Disruptive Event radiological acceptance criterion during the postclosure period.

I. CONCEPTUAL MODEL

The recent illustrative postclosure safety assessment for a used fuel repository located in the sedimentary rock of the Michigan Basin[1] considers gas generation and transport, which can affect both the internal repository pressure and the transport of radionuclides.

Modelling of the internal repository pressure is discussed separately.[2]

The generation and migration of gas is a coupled process, in which gas generation is dependent on the availability and transport of groundwater near the container. Gas can migrate through the repository by:

- *Dilational flow*, where gas pressure exceeds the local confining stress and physically displaces clay-based sealing materials, with gas moving through the resultant open pathway. Pathways are typically small-scale and are localised. They will generally propagate until sufficient pathway volume has been opened to reduce the pressure. Dilational flow paths are typically unstable and will close once the pressure is reduced below the confining stress.

- *Conventional two-phase flow*, where gas pressure is sufficient to partially or completely displace water from the pore space of a material, allowing gas to travel through the connected porosity.

- *Dissolving in groundwater*, where dissolved gas can travel with groundwater flow or diffuse through groundwater.

I.A. Gas Generation

In low-permeability sedimentary rock, saturation of the repository may take thousands of years. This leads to the definition of the following four phases in the evolution of the corrosion environment:

- *Phase 1*: This is an early aerobic period that occurs prior to the onset of aqueous corrosion, immediately

following closure of the repository. In this phase, saturation of the materials surrounding the container has not yet occurred, no liquid water is available for corrosion, and the material immediately adjacent to the container remains unsaturated due to high temperatures. Oxygen is initially present in the unsaturated pore space. If relative humidity is also low, corrosion is limited to slow air oxidation.

- *Phase 2*: This is an unsaturated aerobic phase that occurs following the condensation of liquid water on the steel surface. Steel corrosion is higher under these conditions.

- *Phase 3*: This corresponds to an unsaturated anaerobic phase that occurs following the consumption of oxygen but prior to full saturation of the materials surrounding the container. Corrosion during this period is supported by the cathodic reduction of water accompanied by the evolution of hydrogen.

- *Phase 4*: This is a saturated anaerobic phase that is entered once the materials surrounding the container have become fully saturated by groundwater. Phase 4 corrosion assumes a source of carbonate is present in the neighbouring sealing materials, generating iron carbonate which provides substantially less corrosion protection. As with Phase 3, corrosion during Phase 4 is supported by the cathodic reduction of water accompanied by the evolution of hydrogen.

These four phases do not necessarily occur sequentially. Phases 1 and 2 can both occur under aerobic conditions and the degree to which each of the corrosion processes are active depends on the relative humidity. The Phase 3 and 4 corrosion processes proceed under anaerobic conditions and may occur concurrently. The Phase 3 process also depends on relative humidity.

There are a number of processes, such as methanogenesis, through which the quantity of gas in the repository may be reduced.[3] The gas modelling presented here takes no credit for these processes. Parameter values used in the corrosion calculations are discussed in Ref. 1.

Additional gas sources, such as those arising from corrosion of rock bolts, degradation of trace organic materials in the backfill, and radiolysis of water are neglected on the basis that the amount of gas produced by such processes is much less than the amount of gas produced due to corrosion of all containers.

I.B. Gas Migration and Transport

In this assessment, conventional two-phase flow and groundwater dissolution processes are considered sufficient for estimating gas migration through the repository. This approach is consistent with other programs in which numerical modelling of gas migration has focussed primarily on conventional two-phase flow.[4,5,6]

Two-phase flow gas migration modelling is based on the van Genuchten model for water retention.[7,8] Parameter values are provided in Ref. 1.

Dissolution of gas in water is described by Henry's law, where the concentration of dissolved gas is proportional to the partial pressure of the gas. Salinity reduces the Henry's law coefficient, and a value of 4×10^{-11} mol fraction/Pa is used for hydrogen in this study.[9]

II. COMPUTER CODE

The conceptual model is numerically represented in the T2GGM computer code[3], version 3.1. T2GGM assesses the coupled behaviour between gas generation, temperature, and the movement of gas and water. It is composed of two coupled models: the Gas Generation Model (GGM) used to describe the generation of gas due to corrosion of steel components, and the TOUGH2 model[10] used to describe gas and water transport from the repository and within the geosphere. Key outputs are estimates of the repository pressure, repository saturation, and gas flow rates within the geosphere and repository system.

III. ANALYSIS METHODS

Gas generation and transport are investigated using models at two different scales of resolution: Room-Scale and Repository-Scale.

The Room-Scale Model considers hydrogen gas generation from corrosion processes and two-phase flow with a simplified repository geometry. It also considers thermal effects on groundwater flow and gas flow associated with heat transfer from the used fuel containers. The model domain extends to include a single full placement room and associated cross-cut drift.

Gas behaviour within each placement room is assumed sufficiently similar for Room-Scale results to be representative of gas behaviour in all placement rooms. Gas transport results are scaled to represent entire panels, each compromising either 27 or 28 placement rooms[1], with the scaled results used as input to the Repository-Scale Model. Gas transport data are provided to the Repository-Scale Model at locations where the cross-cut drifts intersect the main access tunnel. A constant

pressure is specified at the cross-cut drift external boundary.

The Repository-Scale Model considers the transport of gas and water, without thermal effects, along the main drifts and shaft of the repository, using results from the Room-Scale Model to estimate the amount of gas reaching the drifts.

III.A. Overall Approach

During setup, the model is initially run for a sufficient period of time to allow the pre-construction conditions to equilibrate. Thereafter, simulations are conducted in three consecutive segments to account for the evolution of repository conditions. These are:

1 *Preclosure* – the engineered facilities are open to atmospheric pressure and are fully saturated with gas for the operational period prior to closure. This is assumed to be seven years for placement rooms and 60 years for the main drift. Desaturation of the neighbouring rock is modelled and used to initialize the postclosure period.

2 *Postclosure / pre-failure* – sealing materials are placed in the repository with specified gas saturations at atmospheric pressure within the room and cross-cut drift (Room-Scale Model) or main drift and shaft (Repository-Scale Model). The system repressurizes and resaturates for 10,000 years with no gas generation taking place.

3 *Post-failure* – the steel container is exposed and gas generation commences.

Pressure, gas saturation and dissolved gas content are continuous within the geosphere from one segment to the next.

The Room-Scale and Repository-Scale models are separate models and manual iteration is performed to ensure consistency between the model pressures at the interface location. A fully integrated model would allow time-dependent calculation of the interface pressure; preliminary results from a prototype integrated model are similar to the results presented here.[2]

III.B. Detailed Transport Models

Within the host rock and the various engineered sealing materials, interactions between gas and liquid are modelled using the van Genuchten equations for water retention.

III.B.1. Room-Scale Model

The Room-Scale Model uses a simplified geometry. The model domain consists of a full placement room and associated cross-cut drift. Gas generation, thermal effects, and two-phase flow are simulated, with gas flow rates in the cross-cut drift calculated for input to the Repository-Scale Model.

The domain includes all 50 containers, the concrete and bentonite seals, and the access drift connecting it to the cross-cut drift. Horizontal symmetry is assumed and only one-half of the cross-section is modelled, including 10 m of host rock horizontally adjacent to the room. The plan-section domain of 514.4 m × 10 m represents the full length of the single room located in the middle of the repository including one-half the intact rock separating the room from adjacent repository panels. The vertical extent of the model is from 1000 metres below the repository to the top of the Devonian formations, 500 metres above.

The 50 containers in the room are grouped into 10 combined containers where each combined container has the corrosion and heat generating characteristics of 5 individual containers. The combined containers have the length and volume of 5 individual containers so that the amount of bentonite is the same as in the actual system. Figure 1 illustrates the model.

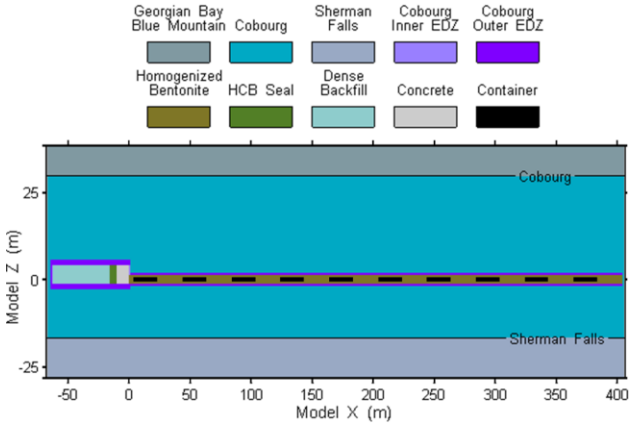

Fig. 1. The Room-Scale Model

Pressure boundary conditions at the top and bottom of the model are set to simulate a system at hydrostatic equilibrium. Variations in fluid density will have negligible impact on the simulations, where pressures and flows are largely driven by processes occurring within the placement room; consequently, initial pressures for all intact rock nodes are set to freshwater hydrostatic.

Only heat generated by radioactive decay is considered. Although aerobic corrosion of steel is exothermic, the total heat generated would be negligible compared to the decay heat. Anaerobic corrosion of steel is endothermic. The heat generated per container was multiplied by 50 to account for the 50 containers in the room, and then divided by two to reflect the half-room geometry. The resulting thermal source term was uniformly distributed among each of the two nodes in each of the ten combined containers.

Because the Room-Scale and Repository-Scale Models are each run independently, a fixed-pressure boundary node is used to couple the Room-Scale Model to the Repository-Scale Model. The boundary node is situated within the cross-cut drift; the node pressure is fixed at 6.8 MPa, determined by a preliminary series of manual iterations of the two models.

Temperature at the boundary node is set to 14°C, determined by preliminary runs to be the long-term average temperature after 100,000 years. The fixed-temperature value is an approximation to simplify implementation. This affects the temperature in the drift, but has only a very small effect on the average temperature in the placement room.

III.B.2. Repository-Scale Model

The Repository-Scale Model considers the transport of gas and water, without thermal effects, along the main drifts and shaft of the repository, using results from the Room-Scale Model to represent the amount of gas reaching the drifts. The model does not simulate any placement rooms.

The model domain consists of a single drift connecting the main and ventilation shafts (see Figure 2). Horizontal symmetry is assumed; half the shafts are modelled. The model extends vertically approximately 400 metres, from 100 metres below the repository.

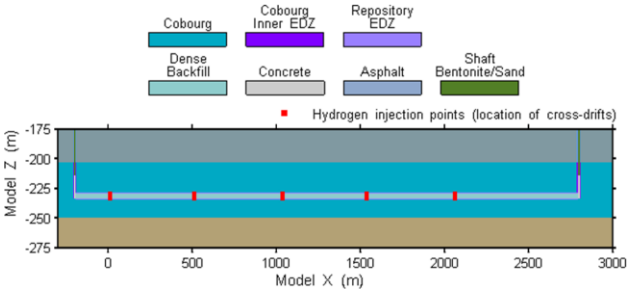

Fig. 2. The Repository-Scale Model

Pressure-boundary conditions in the top and bottom layers of the model are calculated hydrostatic pressures.[1] The remaining sides of the model are no-flow boundaries.

Gas is injected into the model at five locations (see Figure 2), corresponding to intersections of the main access tunnel and the cross-cut drifts. The gas source term is calculated from results of the Room-Scale Model, multiplying the gas flow exiting a single room by the average number of rooms in each panel. This source term ignores the migration and accumulation of gas in the cross-cut and perimeter drifts so that all gas leaving the Room-Scale Model is assumed to immediately reach the main access tunnel.

IV. RESULTS OF GAS GENERATION AND TRANSPORT MODELLING

IV.A. Room-Scale Model

Results from the Room-Scale Model simulation of steel corrosion are shown in Figures 3 and 4. Figure 3 shows corrosion results in terms of container mass remaining (per container); the right-hand axis shows the container mass remaining in terms of container wall thickness.

Figure 4 shows corrosion rates by process. The initial period of Phase 4 corrosion is caused by the high average liquid saturation within the sealing materials at the time of assumed failure. Phase 4 corrosion continues until the average gas saturation reaches 10%, after which only Phase 3 corrosion occurs. The corrosion rate decreases as temperature declines. Gas saturations near the container are relatively constant and remain high enough (greater than 10%) so that resumption of Phase 4 corrosion does not occur until approximately 85,000 years. The container mass is exhausted by 440,000 years.

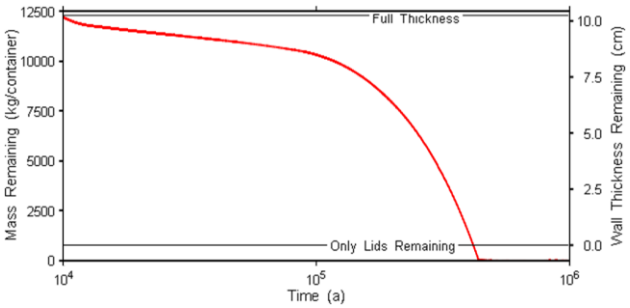

Fig. 3. The Room-Scale Model – Steel Consumption

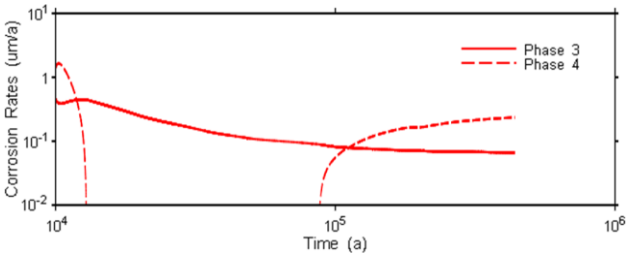

Fig. 4. The Room-Scale Model – Corrosion Processes

Figure 5 presents information on the gas distribution. In general, nearly all generated gas leaves the Room-Scale Model through the cross-cut drift boundary. Of the gas retained, the placement room sealing materials contain the bulk of the generated gas, with the EDZ also containing a significant component for the first half of the simulation. The difference between the dashed line and the topmost solid line indicates the amount of gas that has exited the model domain and entered the Repository-Scale Model.

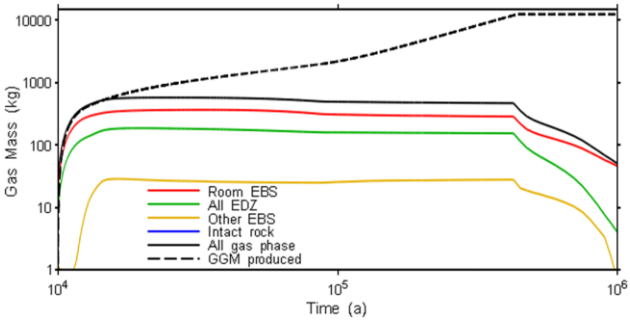

Fig. 5. The Room-Scale Model – Gas Distribution

Figure 6 shows the gas generation rate within the placement room (red line) and the gas flow rate out of the placement room through the drift (blue line).

The placement room buffers the initial pulse of gas until approximately 16,000 years, at which time gas begins to flow out of the model. The onset of Phase 4 corrosion (see Figure 4) is responsible for the change in slope that occurs just prior to 100,000 years. The Phase 4 period conservatively assumes that there is sufficient carbonate available to allow the reaction to proceed unabated (otherwise corrosion and gas generation would continue but at a slower rate). Gas flow ceases at 440,000 years when container mass is exhausted.

IV.B. Repository-Scale Model

Figure 7 shows the total amount of free-phase hydrogen gas within the Repository-Scale Model. Hydrogen gas is found mostly within the engineered sealing materials in the repository tunnels and shafts; however, significant amounts are also within the EDZ. Very little is in the host rock. The onset of Phase 4 corrosion (see Figure 4) is responsible for the change in slope that occurs just prior to 100,000 years.

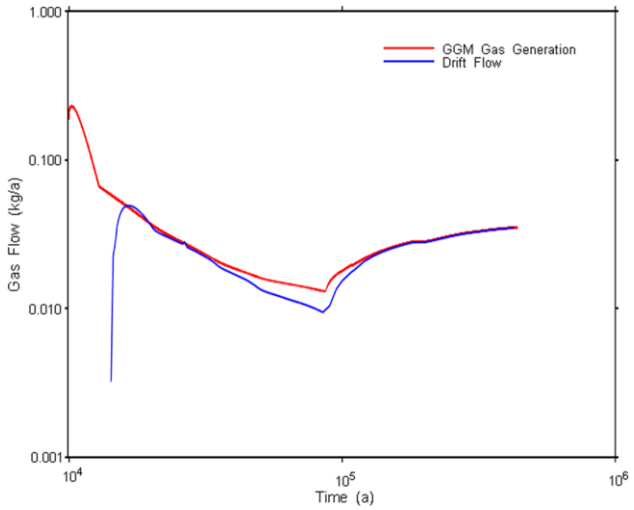

Fig. 6. The Room-Scale Model –Gas Flow Rates

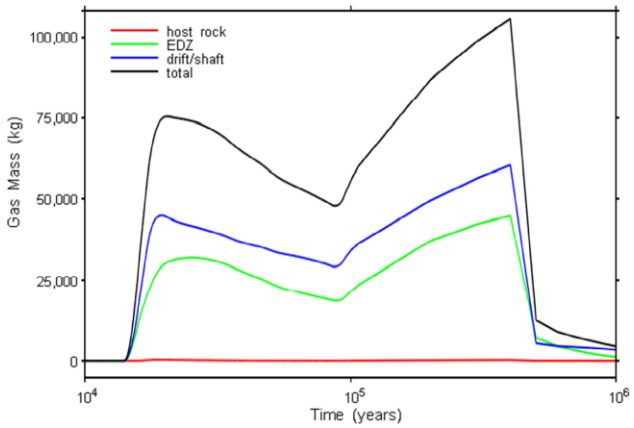

Fig. 7. The Repository-Scale Model – Distribution of Free-Phase Gas

Flow results shown in Figure 8 illustrate the amount of hydrogen gas leaving the top of the model. The grey line represents the total rate at which gas enters the model from the placement rooms while the black line represents the total rate at which gas leaves the model via the shafts and, to a much lesser degree, the rock. The other colours indicate the specific pathways used by the gas to exit the model. Because the Guelph formation (the top of the formation is situated approximately 130 metres below the ground surface) has a much lower gas entry pressure than the shaft sealing materials, the rising hydrogen gas will exit the shafts and enter the Guelph formation.

Model results show that hydrogen is moving primarily in gaseous form, with dissolved hydrogen accounting for approximately 0.01% of hydrogen transport.

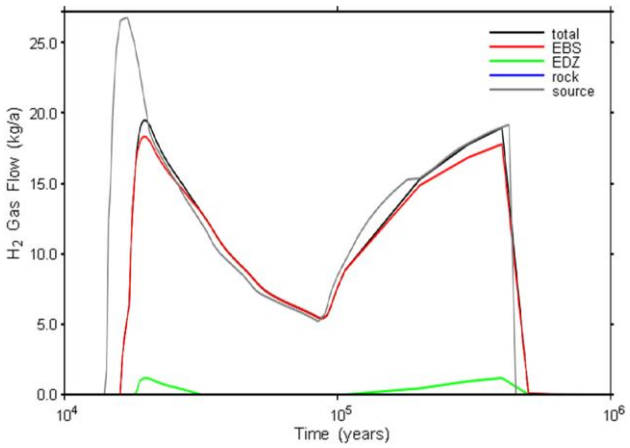

Fig. 8. The Repository-Scale Model – Gas Flow Out of the Top of the Model

V. DOSE CONSEQUENCES

Of the radionuclide species present within the used fuel, very few are volatile or semi-volatile under repository conditions. Of these, only C-14, Cl-36, Se-79, and I-129 have half-lives sufficiently long to be of potential concern.[1,9]

Table I shows the source term for each radionuclide together with their inhalation dose coefficients and Henry's law constants. The products of these three values are also shown to allow a comparison of the relative hazard. Because the product for C-14 is about four orders of magnitude greater than the others, only C-14 is considered further in the dose calculations.

TABLE I. Comparison of Volatile, Long-lived Radionuclides

Nuclide	Half-Life (a)	Source, 10,000 years (Bq/kgU)	Inhalation Dose Coefficient (Sv/Bq)	Henry's Law Constant (C_{gas}/C_{aq})	Product (Sv/kgU)
C-14	5700	4.7×10^{5}	6.2×10^{-12}	10^{2}	2.9×10^{-4}
Cl-36	301,000	1.8×10^{4}	7.3×10^{-9}	10^{-6}	1.3×10^{-10}
Se-79	295,000	1.9×10^{4}	6.8×10^{-9}	10^{-4}	1.2×10^{-8}
I-129	15,700,000	2.1×10^{4}	3.6×10^{-8}	10^{-4}	7.5×10^{-8}

For this assessment, gas-borne dose calculations assume that, upon container failure, the instant-release fraction of C-14 partitions into the gas phase; also, congruent release of C-14 during corrosion of the fuel and cladding is taken into account. This release is assumed to be well-mixed and evenly distributed throughout all of the hydrogen gas in the repository. No credit is taken for loss of C-14 during transport from the placement room to the biosphere, due to chemical reaction, microbial reaction, sorption, or partitioning into groundwater; however, radioactive decay of C-14 is accounted for.

The Repository-Scale Model results presented in Figures 7 and 8 provide the data needed to estimate the rate at which C-14 activity leaves the repository. Figure 7 shows the total amount of hydrogen gas in the repository while Figure 8 shows the rate at which hydrogen moves up the shafts.

As noted above, because the Guelph formation has a much lower air-entry pressure than the shaft sealing materials, the rising hydrogen gas will exit the shafts and enter the more permeable Guelph formation. Once in the Guelph formation, the gas will most likely disperse and dilute laterally underground, so there is no significant vertical flux upwards and no dose consequence to the critical group. Figure 9 shows the rate at which C-14 activity would enter the Guelph formation, taking into account radioactive decay of C-14.

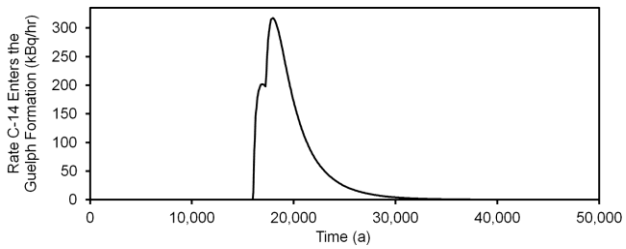

Fig. 9. Rate at which C-14 enters the Guelph Formation

A bounding estimate of the dose consequence can be conservatively obtained by assuming the gas is not dispersed in the Guelph formation and the critical group is exposed. This is done by assuming that all C-14 exits the repository via a single shaft, and that all C-14 in the shaft remains in the shaft until it reaches the surface. After reaching the surface, the C-14 then enters a house that is built directly on top of the shaft, exposing the occupants to an inhalation hazard. Under these assumptions, the rate information in Figure 9 also represents the rate at which C-14 activity enters the house. The maximum value is 320 kBq/hr.

The peak inhalation dose rate to the occupant is determined using data in Ref. 11. For a house volume of 228 m³, an air exchange rate equivalent to a volume fractional turnover rate of 0.35 per hour (i.e., 7×10^{5} m³/a), a dweller inhalation rate of 8400 m³/a, an occupancy

factor of 0.8, and an adult inhalation dose coefficient for C-14 (as CO_2) of 6.2×10^{-12} Sv/Bq (Ref. 12), the peak dose rate is 0.17 mSv/a occurring at 18,000 years.

VI. CONCLUSIONS

Exposure of the steel components of an engineered barrier system to groundwater will result in the generation of gas due to corrosion processes. This gas can affect both the internal repository pressure and the transport of gaseous radionuclides.

A conservative assessment of the dose consequences of gas generation is determined by considering the variant case of the All Containers Fail Disruptive Scenario in which the container failures are all assumed to occur at 10,000 years. In this case, the steel in all containers is exposed and starts to corrode.

Even with the extreme conservatism of assuming simultaneous failure of all containers at 10,000 years, gas-borne dose consequences reach a peak dose rate of 0.17 mSv/a, which is a factor of six below the interim Disruptive Event acceptance criterion of 1 mSv/a. The peak occurs at 18,000 years. For a more realistic case of failure of copper containers over longer times, the dose rates would be lower. For example, if the copper containers all fail on time scales associated with the next glaciation or later, the associated dose rates would be well below 1 µSv/a due to decay of C-14.

ACKNOWLEDGMENTS

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