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Heat Generation and Heating Limits for the IRUS LLRW Disposal Facility

Limite de dégagement de chaleur et d'écchauffement de l'installation IRUS de stockage permanent de déchets radioactifs de faible activité

R.E. Donders, J. Slaby, F. Caron

Paper presented at the 1995 Waste Management Conference
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Chalk River Laboratories
Atomic Energy of Canada Limited
Chalk River, Ontario, Canada K0J 1J0
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and

[*] Mechanical and Aerospace Engineering
Carleton University
Ottawa, Ontario, Canada K1V 6N2
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LIMITE DE DÉGAGEMENT DE CHALEUR ET D'ÉCCHAUFFEMENT DE L'INSTALLATION IRUS DE STOCKAGE PERMANENT DE DÉCHETS RADIOACTIFS DE FAIBLE ACTIVITÉ

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RÉSUMÉ

Le dégagement de chaleur résultant de la désintégration radioactive et de la détérioration chimique des déchets doit être pris en considération pour la mise en oeuvre d'une installation de stockage permanent de déchets radioactifs de faible activité. Ceci est particulièrement important dans le cas de la gestion des sources usées de radio-isotopes. Des calculs sont présentés pour la température et d'échauffement pour l'installation IRUS (installation souterraine anti-intrusion) envisagée pour le stockage permanent à faible profondeur. Les calculs de transfert thermique ont été réalisés à l'aide d'un code utilisant la méthode par éléments finis, à partir de paramètres de transfert thermique et de conditions limites qui sont réalistes, mais prudents. La température de ramollissement des déchets contenant du bitume (38 °C) est le facteur qui limite le degré de dégagement de chaleur dans l'installation. Cela limite aussi le dégagement de chaleur de l'installation IRUS à 0,34 W/m^3, en supposant une source uniforme. Pour une limite de dégagement de chaleur d'ensemble plus basse, on peut accepter des colis qui peuvent dégager une température plus élevée en imposant des restrictions sur leur emplacement dans l'installation. Pour la plupart des déchets radioactifs de faible activité, le dégagement de chaleur résultant de la désintegration radioactive et de la détérioration chimique représente une faible fraction des limites d'échauffement de l'installation IRUS. Toutefois, des restrictions quant à l'échauffement admissible auront des effets sur le stockage permanent des sources radioactives de plus haute activité. Des sources de ^{60}Co de haute activité exigeront des périodes de refroidissement d'environ 70 ans, et certaines sources de ^{137}Cs devront être stockées en permanence dans des installations conçues pour les déchets dégageant une chaleur plus élevée.

Laboratoires de Chalk River
Énergie atomique du Canada limitée
Chalk River (Ontario) K0J 1J0

* Génie mécanique et aérospatial
Université Carleton
Ottawa (Ontario) Canada K1V 6N2

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ABSTRACT

Heat generation from radioactive decay and chemical degradation must be considered when implementing low-level radioactive waste (LLRW) disposal. This is particularly important when considering the management of spent radioisotope sources. Heating considerations and temperature calculations for the proposed IRUS (Intrusion Resistant Underground Structure) near-surface disposal facility are presented. Heat transfer calculations were performed using a finite element code with realistic but somewhat conservative heat transfer parameters and environmental boundary conditions. The softening-temperature of the bitumen waste-form (38°C) was found to be the factor that limits the heat generation rate in the facility. This limits the IRUS heat rate, assuming a uniform source term, to 0.34 W/m^3. If a reduced general heat-limit is considered, then some higher-heat packages can be accepted with restrictions placed on their location within the facility. For most LLRW, heat generation from radioactive decay and degradation are a small fraction of the IRUS heating limits. However, heating restrictions will impact on the disposal of higher-activity radioactive sources. High activity ^{60}Co sources will require decay-storage periods of about 70 years, and some ^{137}Cs will need to be disposed of in facilities designed for higher-heat waste.

Chalk River Laboratories
Atomic Energy of Canada Limited
Chalk River, Ontario, Canada   K0J 1J0

* Mechanical and Aerospace Engineering
Carleton University
Ottawa, Ontario, Canada   K1V 6N2

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1. INTRODUCTION

Heat generation from radioactive decay and chemical degradation must be considered when implementing low-level radioactive waste (LLRW) disposal [IAEA 1985]. This is particularly important when considering the disposal of radioisotope sources. This paper summarizes heating calculations that have been performed for the proposed Intrusion Resistant Underground Structure (IRUS) disposal facility. Heat limits are proposed for the IRUS facility, and these are compared with radioactive decay and chemical degradation heating rates expected in IRUS waste. The implications of heating considerations on managing Canada's LLRW are also discussed.

The IRUS disposal facility (Figure 1) is based on an open-bottom concrete vault, and is designed for radioactive waste with a hazardous lifetime of less than 500 years [Hardy, et al., 1988]. The facility is expected to be filled with higher-activity LLRW from Canadian nuclear research programs and radioisotope applications. The basic design principles for IRUS are to contain the waste within the vault (i.e., restrict the release of contaminants), and isolate the waste from the environment (i.e., prevent inadvertent intrusion).

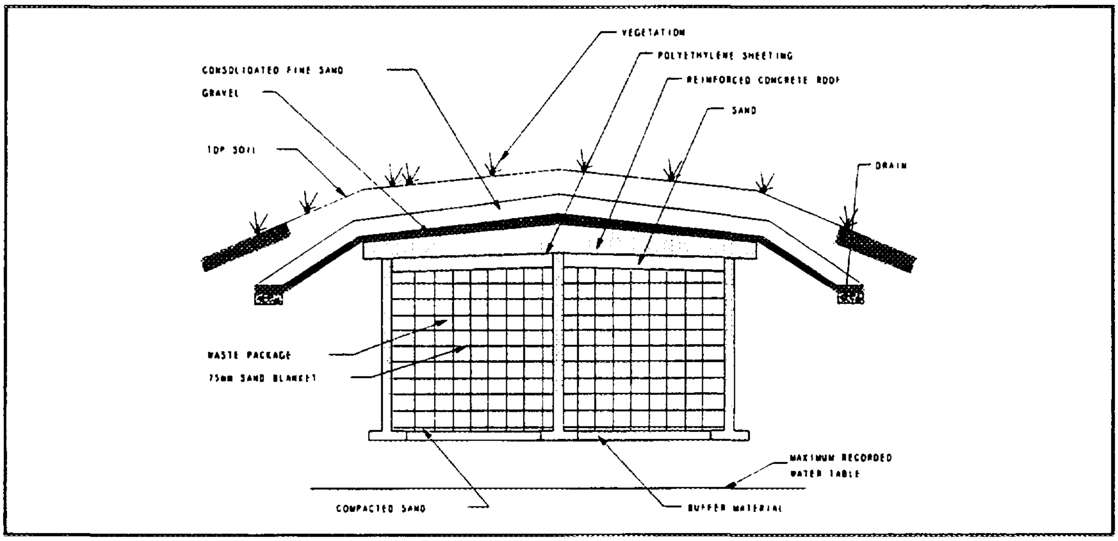

Figure 1. IRUS Facility After Closure.

The IRUS facility is 32 m long, 22 m wide and 9 m deep, and is divided into 6 cells that provide a total useable volume of about 3900 m^3. The reinforced concrete walls are arched so that when the cells are empty, the external soil pressure creates compressive stresses within the walls, reducing the need for reinforcing bars. The walls are 0.61 m thick, and the reinforced concrete roof is 1 m thick. The floor is permeable to avoid the "bathtub effect", and is composed of two buffer layers: a 0.3 m thick mixture of sand (90%) and clinoptilolite (10%), and a 0.3 m thick mixture of sand (90%) and Dochart clay (10%). The clinoptilolite and clay have the capacity to sorb many critical radionuclides from aqueous solution, and thus reduce radionuclide escape from the vault. To avoid flooding, IRUS is to be located in a free draining sand deposit with its foundations at least one meter above the highest recorded water table. Final closure of the facility will include a multilayer earthen cover system about 2 meters thick.

During the operating phase, the IRUS facility will be covered with a temporary weathershield building. The building will contain a gantry crane for transferring materials to the cells using remote attach/detach capabilities so that entry into the cells is not required. The principal waste packages will be 0.4 m^3 bales of compacted waste (90% volume) and 200 litre metal drums. The bales will include fibrous materials, plastics, and small quantities of metals. Most drums will contain a bitumen waste-form produced from liquid-solidification processing or ash immobilization. Waste will occupy about 50 percent of the IRUS volume. Voids between packages and layers of waste will be backfilled with sand (90%) and clinoptilolite (10%).

2. FACTORS LIMITING HEATING RATE IN THE IRUS FACILITY

Factors that could limit the heat generation rate within the facility were determined, as summarized in Table 1. For each factor a maximum allowable temperature or temperature gradient was determined (column 3). The melting temperature of bitumen (38°C) was identified as the factor that would limit the allowable temperature, and thus the heat load, in the IRUS facility.

Table 1. Factors That Could Limit Heating in the IRUS Facility.

Factor	Effect	Temperature or δt
Concrete strength under sustained temperature	Decreased concrete strength	< 38°C - no adverse effect At 75°C - 10-15% decrease
Thermal stress in IRUS structure	Cracking/spallation of concrete	Allowable ΔT = 26.7°C/m
Melting temperature of bitumen	Possibility of settling of particles	38°C
Flash point temperature of bitumen	This factor can be rejected	163°C
Microbiological degradation of bitumen	Generation of gases and degradation of waste form	0.7 mm/100 years under the most favourable conditions, temperature from 0 to 70°C
Gas generation and other biogeochemical reactions	This factor can be rejected since the waste is surrounded by sand/clinoptilolite	
Tritium migration	Contamination	Needs to be determined
Container corrosion	Containers are not credited as a barrier to contaminant migration	

Concrete strength is generally not affected by temperatures less than 38°C [Mittelacher, 1992]. At temperatures above 75°C, decreases in strength have been measured by Carette and Malhotra [1985]. Thermal stresses due to temperature gradients in the concrete walls during the operational and post-closure phases have been considered for the temperature design load of the IRUS structure [Mok, 1989]; the largest temperature gradient of the study was assumed to be a maximum allowable temperature gradient in the concrete for this analysis (i.e., 26.7°C/m). The waste-form most sensitive to increased temperatures is the bitumen used for liquid solidification. This bitumen is similar to type 80/100-150, which has a melting point in the range of 38-53°C [IAEA, 1993]. Bitumen's flash point temperature is well above its melting temperature, and is not a limiting factor. Microbiological degradation of hydrocarbons in bitumen occurs at temperatures from 0 to 70°C. Under the most favourable conditions of temperature, pH, and O_2 availability for degradation, the maximum microbial penetration of bitumen would be of the order of 0.7 mm per 100 years [Allison, et al., 1991]; therefore, it is not expected to limit the temperature in the IRUS facility. Temperature gradients in the waste will increase tritium migration. The allowable temperature gradient for tritium migration have not been determined, but this consideration is not expected to be critical. Finally, the steel containers of the waste are not given any credit as a barrier to waste migration, so corrosion rate temperature dependence will not limit the allowable temperatures.

3. TEMPERATURE CALCULATIONS FOR THE IRUS FACILITY

The finite element computer code, ANSYS 5.0A, was used to perform calculations of temperature in and around the IRUS facility. Calculations were performed for a base case with uniform heating throughout the facility, and for a case where a limited number of high-activity packages were included. Analyses of sensitivity to the various thermal conductivities, boundary conditions and location of facility with-respect-to surface and aquifer were also performed. A 2-D cross-section of the two middle cells of the repository, where the temperature would be the highest, was used as a basis for the simulation. The cross-section was simplified, as shown in Figure 2, using the following assumptions:

- All cells have the same heat load, giving plane symmetry in the centre of the inner concrete wall.
- The waste was assigned a uniform thermal conductivity based on thermal conductivity of building rubble materials, fibrous materials, plastics, sand, and bitumen.
- The overburden layers (gravel, cobblestone, fine sand, and top soil) and buffer layer (90% sand, 10% clinoptilolite) were assigned the thermal conductivity of sand. This is justified since the thermal conductivities of these layers are similar, and minor differences in their thermal conductivities will not significantly impact the calculated temperatures.

3.1 Thermal Conductivities

Typical values and bounding ranges were identified for the thermal conductivities required for the temperature simulations (Table 2). The bounding ranges were used for sensitivity studies, and to help establish a conservative reference scenario.

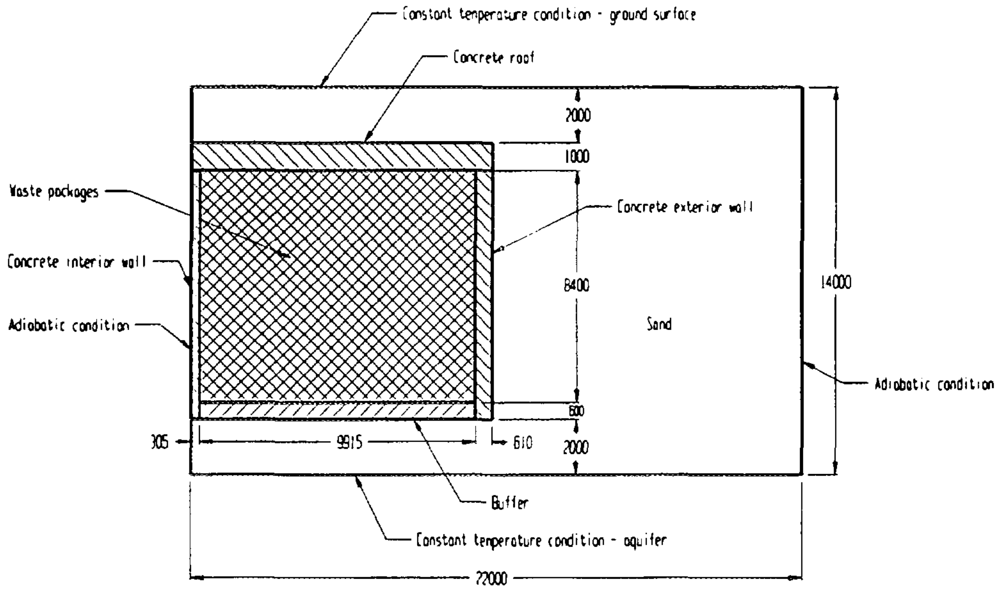

Figure 2. Sketch of the ANSYS Model of the IRUS Facility

Table 2. Parameters Determined for the Analysis

Material	Units	Typical value	Lower bounding value	Upper bounding value
k_{waste}	(W/m.K)	0.17	0.08	0.7
$k_{concrete}$	(W/m.K)	1.56	1.2	1.8
$k_{sand/buffer}$	(W/m.K)	1.4	0.4	2.1
T_{ground}	(°C)	7.9	0.8	14.4
$T_{aquifer}$	(°C)	8.1	3.1	13.1

The walls and the roof of the IRUS facility will be constructed of high-performance reinforced concrete with limestone aggregate. The typical thermal conductivity of concrete with limestone aggregate is 1.56 W/m.K; the selected range of 1.2 to 1.8 W/m.K is slightly more conservative than the typical range. The thermal conductivities of the sand, overburden and buffer is typical of Ottawa area sand with a moisture content of 2-4%. The effect of moisture migration on thermal conductivity of sand was also considered; it was concluded that the selected range of the thermal conductivity of sand covers the effect of moisture migration satisfactorily.

The thermal conductivity of the heterogeneous waste/backfill mixture expected within IRUS has not been explicitly determined. The dry environment within the vault, coupled with the slightly elevated temperatures expected, suggest that the thermal conductivity of dry sand (0.35 W/m.K) would be a reasonable starting point. Fibrous materials and air gaps in the compacted bales may decrease this value somewhat. The thermal conductivities selected for this study cover the range of realistic values, although the "typical value" is likely to be somewhat low.

3.2 Boundary Conditions

The boundary conditions were established based on annual averages of environmental parameters. Annual averages of environmental parameters were used throughout this study, since seasonal temperature variations are insignificant two or three metres below the ground surface [Wildsmith, 1976]. Parameters such as air temperature, ground temperature, aquifer temperature, solar radiation, wind speed and ground-to-sky radiation for the Chalk River/Ottawa Valley region were considered. Since the radionuclides in the IRUS facility will be active for several decades, slightly conservative ranges of annual averages of all parameters were used, based on measured data.

Adiabatic boundary conditions were assigned at the vertical boundaries, as shown in Figure 2. The location of the right boundary was sufficiently far away to not affect the temperatures in or around IRUS facility. At the horizontal boundaries, constant temperature boundary conditions were assigned. The ground surface temperature range was based on the sensitivity analysis of the environmental parameters; therefore, it covers the effects of all those parameters on the ground surface temperature. The effect of heating in the IRUS facility on the ground surface temperature was also examined. It was found that heat load of 2 W/m^3 (which is about 5.5 times higher than the established limit) affects the ground surface temperature only by 0.2 to 0.3°C.

3.3 Effect of Voids Within the Vault

Voids may form within the vault, either through incomplete backfilling around waste or waste slumping. The effects of voids on the heat transfer characteristics were investigated. Horizontal air gaps up to 0.5 m were considered. In the ANSYS model, the closed air gap is represented as a solid material having an effective thermal conductivity of the air gap [Holman, 1981], which includes the effects of conduction, convection and radiation. The effective thermal conductivities, found for various air gap heights for a heat load corresponding to the temperature limit of 38°C in the waste, were used in the finite element simulation.

For vertical air gaps, it was found that the effective thermal conductivities were within the bounding values of the thermal conductivity of waste, and in some instances slightly higher than, therefore, their thermal effect is effectively covered by the waste thermal conductivity.

3.4 Calculations

The problem of the IRUS heating was simplified to five parameters, as listed in Table 2. Temperature calculations were performed to determine the heat generation rate which produces the limiting temperature of 38°C. Calculations were made for a typical case with uniform heat generation throughout the waste, and then single parameter sensitivity analyses were carried out to lower and upper bounding values.

Various schemes were considered that allowed some higher-activity packages to be placed in IRUS. Such schemes must be simple to implement operationally, and they must be amenable to simulation. A suitable scheme reduces the general heat limit for waste, but allows for higher-activity packages to be placed anywhere within 1.2 m of the top or bottom of the facility. To simulate higher activity packages, some finite elements were assigned higher heating rate than others according to selected perturbation schemes (i.e., higher heating in top and bottom layers).

4. RESULTS

The calculated heat rate limits for IRUS that give a maximum temperature of 38°C are presented below. In all cases the temperature gradients in the IRUS structure are less than 50% of the imposed 26.7°C/m limit.

4.1 Heating Rate Limit With Uniform Heat Source

Calculations using the typical values for heat transfer parameters given in Table 2, and assuming uniform heat generation throughout the waste, give an IRUS heat rate limit of 0.63 W/m^3. Single parameter sensitivity analysis showed that the heat limit is not very sensitive to the parameters in Table 2 except for the k_{waste}. The heat load is, in most cases, in the range of 0.5 to 0.7 W/m^3 except for the lower bounding value of k_{waste} when it is 0.35 W/m^3. Analysis also showed that a 1 m increase in the distance between the IRUS facility and the aquifer or ground surface decreased the heat load by about 0.02 W/m^3. Due to uncertainty in some of the parameters, and recognizing that the IRUS facility will need to maintain its performance for several centuries, the reference case was defined as having a typical thermal conductivity of waste form and conservative bounding values of all the other parameters. This conservative reference case gives an IRUS heat rate limit of 0.34 W/m^3. If the waste form thermal conductivity can be better defined, then the heat limit can be adjusted using the data in Table 3.

4.2 Heating Rate Limit With Higher Activity Package Perturbation

The higher activity package perturbation calculations were performed using the same reference case boundary conditions and thermal conductivities as described earlier for the uniform heating source. The calculations show that IRUS could handle a small number 0.4 m^3 packages with heating rates of about 10 W/m^3. If a general heat limit of 0.1 W/m^3 is applied to the facility, then high-activity packages with heating rates up to 1.6 W/m^3 could be placed anywhere within 1.2 m of the top or bottom of the facility.

Table 3. Heat load limit for a uniform heat source, as a function of waste thermal conductivity.

Thermal conductivity of waste (W/m.K)	**Heat load limit (W/m^3)**
0.08	0.22
0.17	0.34
0.5	0.57
0.7	0.63

5. HEATING WITHIN IRUS

Heating in LLRW occurs from radioactive decay and chemical reactions during the waste degradation process. In general, heating in LLRW is expected to be small, although some radioactive sources do produce significant power.

5.1 Radioactive Heating

In solid-LLRW, all radioactive decay energy, except neutrino energy, is deposited locally (neutrino interaction cross sections are negligible). The heat energy and corresponding Bq/Watt for some representative radionuclides in LLRW are listed in Table 4, columns 2 and 3, respectively. These values were derived from average energy released per decay provided in ICRP Publication 38 [1983].

Table 4. Decay energies for some important radionuclides in LLRW, and IRUS radionuclide limits (for single nuclides) based on heating considerations only.

Decay Chain	**Heat Energy** (MeV)	**Bq/Watt Heat** (Bq/W)	**IRUS Limit based on 0.34 W/m^3** (Bq*/m^3)	**IRUS Limit based on 0.1 W/m^3** (Bq*/m^3)	**IRUS Limit based on 1.6 W/m^3** (Bq*/m^3)
^{3}H → ^{3}He	0.006	1.1E+15	3.7E+14	1.1E+14	1.8E+15
^{14}C → ^{14}N	0.05	1.3E+14	4.3E+13	1.3E+13	2.0E+14
^{36}Cl → ^{36}Ar & ^{36}S	0.27	2.3E+13	7.7E+12	2.3E+12	3.6E+13
^{60}Co → ^{60}Ni	2.60	2.4E+12	8.2E+11	2.4E+11	3.8E+12
^{90}Sr → ^{90}Zr	1.13	5.5E+12	1.9E+12	5.5E+11	8.8E+12
^{137}Cs → ^{137}Ba	0.81	7.7E+12	2.6E+12	7.7E+11	1.2E+13
^{226}Ra → ^{206}Pb**	32.9	1.9E+11	6.4E+10	1.9E+10	3.0E+11
^{241}Am → ^{237}Np***	5.7	1.1E+12	3.8E+11	1.1E+11	1.8E+12

* Activity of decay-chain parent only.

** All daughters assumed to be in secular equilibrium with the parent radionuclide.

*** ^{237}Np is unstable, but its half-life is >5000 times that of ^{241}Am, and the peak heating rate from the ^{237}Np decay chain will be much less than from the ^{241}Am decay.

As presented earlier in Section 4, the heat rate limit in IRUS could be implemented as either a uniform limit of 0.34 W/m^3, or alternately, as a base value of 0.1 W/m^3 with some higher-activity packages permitted with heating rates up to 1.6 W/m^3. Radionuclide limits for these different options are presented in columns 4, 5, and 6 of Table 4. These are single nuclide limits, and a sum-of-fraction rule must be applied to mixtures of radionuclides.

To assess the impact of heating limits on IRUS waste acceptance, the expected LLRW inventory must be considered. Preliminary inventory estimates suggest that the total activity in the IRUS vault will be about 4.0 x 10^{14} Bq, with ^{3}H (1.6 x 10^{14} Bq), ^{137}Cs (1.1 x 10^{14} Bq), and ^{60}Co (2.1 x 10^{13} Bq) being the highest- activity longer-lived nuclides (Hardy et al., 1991). The total activity results in about 40 watts of radioactive heating [Chan et al., 1994], which translates to an average heating value of 0.01 W/m^3. This low value suggests that a general heat limit of 0.1 W/m^3 would be acceptable for IRUS, thus permitting a limited number of packages with heating rates up to 1.6 W/m^3.

The IRUS facility could be used for the disposal of sealed radioactive sources, although heating rate and risk-based limits will exclude acceptance of some sources. Table 5 shows the distribution of activities for the higher-activity sealed sources currently licensed in Canada [Beriault, 1994]. Only sources with a half-life greater than three years are included in the table, since shorter half-life sources can be easily stored until their heat generation rates are low. The final column in the table shows the IRUS activity limit in a 0.25 m^3 package, based on a heat limit of 1.6 W/m^3 and assuming that the waste occupies 50% of the vault volume. Sealed sources that exceed this limit are indicated with shading in the table.

The data in Table 5 show that only ^{60}Co, ^{137}Cs, and ^{241}Am sources are above the IRUS heat limit in significant quantities. The very-high-activity ^{60}Co sources, which represent several 100 kW of power, would need to be stored for a period of about 70 years before they qualify for disposal in IRUS. Although the ^{137}Cs sources represent a much smaller heat source than the cobalt, their longer half-life makes decay-storage impractical. High-activity cesium sources will either have to be dismantled to distribute the heat source, or disposed in a facility that can accommodate higher-heat waste. The higher-activity americium sources are not likely to be suitable for near-surface disposal (excluded by risk-based limit), and these sources will likely be disposed in a geologic facility.

Table 5. Licensed radioactive sealed sources in Canada

Radio-nuclide ($T_{½}$ in years)	**Activity Range**					**IRUS Heat Limit for 0.25 m^3 Package***
	0.37-3.69 E+11 Bq	**3.7-36.9 E+11 Bq**	**37-369 E+11 Bq**	**370-3699 E+11 Bq**	**>3699 E+11 Bq**	
^{3}H (12)	79	37	5	1		8800E+11
^{14}C (5730)			1			1000E+11
^{60}Co (5)	18	89	114	53	63	19E+11
^{63}Ni (100)		1	1			3000E+11
^{85}Kr (11)	169	7	1			200E+11
^{90}Sr (29)	15	1				44E+11
^{133}Ba (11)		1				110E+11
^{137}Cs (30)	649	266	28	37	2	62E+11
^{154}Eu (9)		1				32E+11
^{238}Pu (88)	1	5	1			9.0E+11
^{241}Am (432)	138	20				8.8E+11

* Based on a heat rate limit of 1.6 W/m^3 and assuming 50% waste packing efficiency (i.e.,effectively 3.2 w/m^3)

5.2 Heat Generated from Biogeochemical Processes Associated with Waste Decomposition

Leaching experiments have been performed on compacted bales of LLRW to monitor their degradation. The degradation process can be related with simple redox reactions to eventually give heat generation rates. To extrapolate the leaching measurements to give an estimate of the IRUS heat generation rate, a correction factor must be applied to reflect the drier environment expected in IRUS.

Microbial degradation of organic matter in landfill sites is one of the major sources of heat. This reaction gives the theoretical yield of anaerobic degradation of cellulose [Pirt, 1978; Rees, 1980]:

$$1\ \text{kg}\ C_6H_{12}O_6 \rightarrow 0.25\ \text{kg}\ CH_4 + 0.69\ \text{kg}\ CO_2 + 0.056\ \text{kg dry biomass} + 632\ \text{kJ heat} \qquad \textbf{(1)}$$

The water content and the flora dictate the regime of the reaction (aerobic or anaerobic). In a closed system such as a landfill or a confined aquifer, the most energy-yielding substrates will be assimilated preferentially either until depletion, or until the concentrations of these substrates become too low to be favourable thermodynamically. The substrates are generally reduced in this approximate order: O_2, NO_3^-, Mn(IV), Fe(III), SO_4^{2-}, CO_2, N_2 [Scott and Morgan, 1990]. Several redox couples may coexist, due, in part, to the relative closeness in redox couples and heterogeneities in the reactor.

Four bales of compacted low-level wastes were sealed in separate metal boxes to monitor the decomposition process. Each box was connected to a pump in a closed loop and water was recirculated daily over the bales to keep them wet, but not flooded. Major ions, gases, dissolved organic matter (DOM), etc., were monitored periodically [Caron, 1994] on all the bales. Two complete sets of redox-sensitive parameters were used on two dates (spanning over approximately one year) for mass balance calculations. The parameters showing the most important changes are shown in Table 6 (O_2, SO_4^{2-}, NO_3^- and Mn were measured but their contribution was small). They were related together with a series of redox reactions (Table 7) to determine the contribution of each reaction in that time period. The energy released is given by the enthalpy ΔH (negative values denote exothermic reactions).

Table 6. Main chemical parameters measured in the bale leaching experiments. The results shown are for bale sampling station #6.

Parameter	Concentration* at Day #		Number of Moles at Day #		
	32	390	32	390	Difference (+/-)
Gases					
CO_2	27.5	34.2	3.0	3.54	+ 0.54
CH_4	3.9	20.2	0.43	2.11	+ 1.68
N_2	68.3	45.2	7.48	4.72	- 2.76
Dissolved species					
NH_4^+	Not measured	Not measured	(?)	5.52**	+ 5.52
DIC	9.7	15.5	0.33	0.99	+ 0.66
DOM	924.	4458.	1.31	11.9	+ 10.59
$Fe_{(T)}$	12.89	46.55	0.44	2.98	+ 2.54
pH	4.83	5.82			

* Concentrations: dissolved ions in millimole/L; DOM in mg C/L; gases in volume percent.
** Calculated (see text).

Table 7. Complete set of reactions for mass balance.

Reaction	ΔG^o_r **kJ**	ΔH_r^o **kJ**	**Contribution of Reaction #**
(1) $2\ CO_2(g) + 7\ H+ + 4\ Fe(s) \rightleftharpoons$ $1\ CH_3COO^- + 4\ Fe^{2+} + 2\ H_2O$	-305	-627	0.636
(2) $C_6H_{12}O_6 + 6\ H_2O + 4\ N_2(g) + 8\ H^+ \rightleftharpoons$ $6\ CO_2(g) + 8\ NH_4^+$	-669	-433	0.689
(3) $C_6H_{12}O_6 \rightleftharpoons 3\ CH_3COO^- + 3\ H^+$	-194	-185	3.315
(4) $CO_2(g) + 4\ H_2(g) \rightleftharpoons CH_4(g) + 2\ H_2O$	-130	-253	1.679

5.2.1 Heat Production Calculations

The time "zero" of the experiment was set when water was added to each bale. The results of Table 6 are shown for only one bale as an example. The most important assumptions related to these calculations are:

- the gas generation rates were linear over the duration of the experiment, based on the long bale-degradation half-life (30-2400 years)[Caron et al., 1995];
- all the DOM was acetic acid, which is reasonable based on elevated levels of acetate measured in these bales, [Caron, 1994];
- nitrogen was assimilated to produce ammonia, and not displaced. Ammonia was not measured, but the calculated concentration (~0.08 mole/L) is not unrealistic of landfills [Rees, 1980; Lisk, 1991; Ehrig, 1983].

Only the spontaneous reactions (noted with a negative ΔG°_r value) were used because microorganisms are unlikely to go against an energy expense for fulfilling their energy needs. Hydrogen gas was not detected, but its assimilation (reaction 4 in Table 7) and production are more than likely.

The total energy released to the surroundings is the sum of all the enthalpies of each of the reactions (Table 7):

$$\Delta H_{reaction} = \sum_{i=1}^{n} \left(C_i \times \Delta H_r^\circ\right)_i \qquad \textbf{(2)}$$

where i corresponds to reactions 1 to n, and C_i is the relative contribution of reaction i. This amounts to 1734 kJ for bale #6, which is higher than the energy released (840 kJ) from the stoichiometric reaction (1), corrected for the same amount of cellulose. The major difference between our calculation and equation (1) is the contribution of metal corrosion (reaction 1 in Table 7) and CO_2 reduction to methane (reaction 4 in Table 7). Credit is not given to energy losses due to conversion to biomass.

5.2.2 Degradation Rates Applicable to the IRUS Environment

To estimate the heating rate expected in IRUS, the heating rates derived from the leaching tests must be corrected to reflect the dry environment expected in IRUS. Gas generation rate measurements by Torok and Haas [1992], using the same bales degrading in dry conditions, allow for a reasonable estimate of the required correction factor. Their reference gas generation rate was 0.021 l/kg°a. Gas generation rates in the leaching tests (Table 8) were 5 to 60 times higher than this reference value, so the heat generation was normalized to the same rate to give corrected values.

Table 8. Heat generation from the eight bales, corrected to reflect the degradation rates expected in IRUS.

Bale #	**Gas Generation Rate l/kg°a**	**$\Delta^{\circ}H_r$ kJ**	**Corrected Heat kJ°a^{-1}°m^{-3}**	**Generation Rate W°m^{-3}**
1	0.72	-1603	-47	0.0015
4	1.30	-2574	-42	0.0013
5	0.22	-222	-21	0.0007
6	0.10	-1734	-364	0.0115
Average Value				**0.0038**

6. CONCLUSIONS AND RECOMMENDATIONS

Factors that could limit the temperatures and temperature gradients in IRUS were investigated, and the melting temperature of the bitumen waste form (38°C) was found to be the most restrictive. Heat transfer calculations, based on realistic, although somewhat conservative heat transfer parameters and boundary conditions, suggest that a heat rate limit of 0.34 W/m^3 is appropriate for IRUS, assuming a uniform heat source throughout the facility. Alternatively, a general heat limit of 0.1 W/m^3 could be applied, with heating rates up to 1.6 W/m^3 within 1.2 m of the top or bottom of the facility. Assuming a 50% waste packing efficiency, the heat limits for the waste packages will be double the facility limits. For example, a general package heat limit of 0.2 w/m^3 can be applied, with some high-activity packages accepted with heating rates up to 3.2 w/m^3. Heat limits are sensitive to the waste form thermal conductivity. The proposed heat limits are based on a somewhat conservative waste-form thermal conductivity, and this should be reviewed once the actual IRUS inventory is known.

Heating in LLRW occurs from radioactive decay and chemical reactions during the waste degradation process. For most of the LLRW expected to be disposed of in IRUS, radioactive decay heating is expected to be about 0.01 W/m^3. The IRUS facility may also be used for the disposal of sealed sources. A review of sealed sources licensed in Canada shows that most sealed sources fall within the 1.6 W/m^3 heat limit noted earlier. The exceptions to this are ^{60}Co, ^{137}Cs, and ^{241}Am. ^{60}Co sources could be decay-stored for about 70 years at which time they could be economically disposed of in IRUS. On the other hand, the longer-lived ^{137}Cs and ^{241}Am sources will likely need to be disposed of in facilities that can accommodate higher-heat loads.

Heating generation from waste degradation due to biogeochemical processes has been evaluated based on leaching experiments with compacted waste bales. The results, corrected to account for the dry environment expected in IRUS, suggest that chemical heating will contribute about 0.004 W/m^3, which is a small fraction of the proposed IRUS heat limits.

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