ESTIMATION OF RADIOLYTIC GAS GENERATION RATE FOR CYLINDRICAL RADIOACTIVE WASTE PACKAGES – APPLICATION TO SPENT ION EXCHANGE RESIN CONTAINERS

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ABSTRACT

Radioactive waste packages containing water and/or organic substances have the potential to radiolytically generate hydrogen and other combustible gases. Typically, the radiolytic gas generation rate is estimated from the energy deposition rate and the radiolytic gas yield. Estimation of the energy deposition rate must take into account the contributions from all radionuclides. While the contributions from non-gamma emitting radionuclides are relatively easy to estimate, an average geometry factor must be computed to determine the contribution from gamma emitters.

Hitherto, no satisfactory method existed for estimating the geometry factors for a cylindrical package. In the present study, a formulation was developed taking into account the effect of photon buildup. A prototype code, called PC-CAGE, was developed to numerically solve the integrals involved. Based on the selected dimensions for a cylinder, the specified waste material, the photon energy of interest and a value for either the absorption or attenuation coefficient, the code outputs values for point and average geometry factors. These can then be used to estimate the internal dose rate to the material in the cylinder and hence to calculate the radiolytic gas generation rate.

Besides the ability to estimate the rates of radiolytic gas generation, PC-CAGE can also estimate the dose received by the container material. This is based on values for the point geometry factors at the surface of the cylinder.

PC-CAGE was used to calculate geometry factors for a number of cylindrical geometries. Estimates for the absorbed dose rate in container material were also obtained. The results for Ontario Power Generation’s 3 m^3 resin containers indicate that about 80% of the source gamma energy is deposited internally. In general, the fraction of gamma energy deposited internally depends on the dimensions of the cylinder, the material within it and the photon energy; the fraction deposited increases with increasing dimensions of the cylinder and decreases with increasing photon energy.

INTRODUCTION

Ontario Power Generation (OPG) operates several nuclear reactors at two generating sites in Ontario. The primary heat transport (PHT) and moderator purification circuits at these stations generate intermediate level resin waste. Dewatered resin wastes are shipped for storage in 3 m^3 steel containers.

Radioactive waste packages containing water and/or organic substances have the potential to radiolytically generate hydrogen and other combustible gases such as methane and carbon monoxide. The rate of radiolytic gas generation is an important consideration for the safe management of the resin waste both in the short term, for example, during shipment and in the longer term during extended storage.

Typically, the radiolytic gas generation rate is estimated from the product of the energy deposition rate and the radiolytic gas yield (or G value); the latter is usually given in units of molecules gas per 100 eV of absorbed energy. Estimation of the rate of energy deposition within a radioactive waste package must take into account the contributions from alpha, beta, X-ray and gamma emitters present in the waste. While the energy or dose deposited from alpha, beta and X-ray (low energy) emitting radionuclides in the package is easy to estimate because it is deposited locally within the material, estimation of dose from gamma emitters is not straightforward because of the penetrating nature of gamma radiation.

Typically a point kernel approach is utilised to formulate an expression for the gamma dose rate at an internal point within a package (1). Taking an integral over the volume of the package provides an estimate of the dose rate at the selected point from all volume elements in the package. The expression for the dose rate can be separated into two terms: a constant term outside the integral and all terms dependent on the geometry of the package are collected within the integral. The latter is called a point geometry factor. Because the average dose rate within the package is generally of greater interest than the dose rate at a specific point within the package, an average geometry factor must be estimated by volume averaging the point geometry factor.

Until recently, no satisfactory method existed for estimating the point and average geometry factors for a cylinder. For this reason, assessments of the radiolytic H_2 gas formation rate in Ontario Power Generation (OPG)'s resin containers assumed that, as in the case of energy or dose deposition from alpha, beta and X-ray emitting radionuclides, the energy associated with the decay of gamma emitting radionuclides is also completely deposited within the package. The validity of this assumption was not known.

Therefore, development of a methodology for rigorously estimating the radiolytic gas generating rate from gamma emitters present in a cylindrical waste package was undertaken. As part of this, the theory for estimating the average geometry factor for a cylinder was formulated. This takes into account dose buildup factors which are important for large industrial waste packages such as OPG's 3 m^3 spent resin container. A prototype code (named PC-CAGE) was developed for the numerical solution of the integrals involved. It was implemented in Visual C++ as a Windows based application.

An outline of the theory for estimating the radiolytic gas generation rate, including a generalized treatment for estimating geometry factors, is presented in this paper. Details of the derivation of the geometry factors for a cylinder are being published elsewhere (2). A comparison of geometry factor values based on PC-CAGE with literature-based values is also included here. Finally, previous estimates of the radiolytic gas generation rate in OPG's spent resin containers are compared with estimates based on the calculated geometry factors.

ESTIMATION OF THE TOTAL RADIOLYTIC GAS GENERATION RATE

The radiolytic gas generation rate is generally estimated from the product of the energy deposition rate and the radiolytic gas yield (or G value). Therefore, the problem of estimating the radiolytic gas generation rate reverts to the estimation of the energy deposition rate in MeV/s/m^3 or other equivalent units. As mentioned earlier, the total rate of energy deposition or dose rate $\dot{D}$ equals the dose rate contributions from alpha, beta, X-ray and gamma emitters present in the waste. Formally, this can be expressed as

$$\dot{D} = \dot{D}_\alpha + \dot{D}_\beta + \dot{D}_X + \dot{D}_\gamma \qquad \text{(Eq. 1)}$$

where each dose rate term has units of MeV/s/m^3. The dose rate term corresponding to alpha emissions can be expressed as

$$\dot{D}_\alpha = \sum A\, E_{avg}\, I \qquad \text{(Eq. 2)}$$

where the summation includes the contributions from all alpha emitting radionuclides, and

- A represents the activity of a radionuclide in Bq/m^3,
- E_{avg} represents the average energy emitted (MeV) per disintegration, and
- I represents the number of alpha particles emitted per disintegration.

Similar expressions apply for the calculation of $\dot{D}_\beta$ and $\dot{D}_X$. However, because of the penetrating nature of gamma radiation, $\dot{D}_\gamma$ has the form

$$\dot{D}_\gamma = \sum A\, \{\, I_{E_1} f(E_1) + I_{E_2} f(E_2) + \ldots\} \qquad \text{(Eq. 3)}$$

which may be approximated as

$$\dot{D}_\gamma \sim \sum A\, f(E_{avg})\, I \sim \sum S_v\, f(E_{avg}) \qquad \text{(Eq. 4)}$$

where, as before, the summation is taken over the contributions from all gamma emitting radionuclides, and

- I_{E_i} represent the number of photons of energy E_i emitted per disintegration with E_1, E_2… etc being the energies corresponding to the multiple emissions, if any, from a given radionuclide,
- f is a function of energy that incorporates the geometry factor (discussed in the next Section),
- I represents the total number of gamma photons emitted per disintegration (i.e., $I = I_{E_1} + I_{E_2} + \ldots$) and

S_V equals the product AI and represents the volumetric gamma photon flux, γ/(s m^3).

Estimation of f in Equation (4) is discussed next.

ESTIMATION OF DOSE RATE FROM GAMMA EMITTERS WITHIN A CYLINDRICAL RADIOACTIVE WASTE PACKAGE

Consider the case of a radionuclide which emits a gamma photon with an energy E [MeV] at the rate of S_V [γ/(s m^3)] inside a source volume V_S (see Fig. 1). The dose rate, $\dot{D}_\gamma$ [MeV/(s m^3)] received by a target with volume V_T is then given by the expression (1):

$$\dot{D}_\gamma = \frac{\int_{V_S}\int_{V_T} \frac{S_V dV_S}{4\pi r^2} e^{-\mu r} E \mu_a B(\mu r) dV_T}{V_T} \qquad \text{(Eq. 5)}$$

where μ is the total linear attenuation coefficient (m^{-1}) for gamma photons of energy E, μ_a is the linear energy absorption coefficient (m^{-1}) and $B(\mu r)$ is the buildup factor.

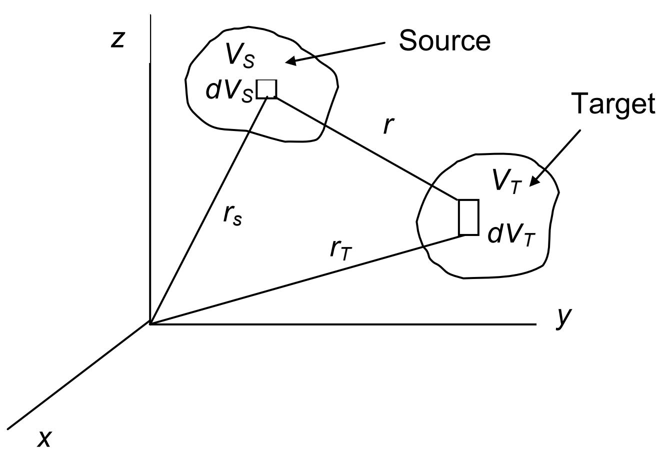


Fig. 1 Geometry for Source and Target (1)

As given by Tsoulfanidis (1), the geometry factor g, defined as

$$g = \frac{1}{V_T}\int_{V_T} dV_T \int_{V_S} \frac{e^{-\mu r}}{r^2} B(\mu r) dV_S \qquad \text{(Eq. 6)}$$

has units of length. Equation (5) can, therefore, be written as:

$$\dot{D}_\gamma = \frac{S_V}{4\pi} E\,\mu_a\,g = S_V f \qquad \text{(Eq. 7)}$$

where f equals $E\,\mu_a g/4\pi$ and is a function of energy (see Equation 4). When the source is also the target (i.e., $V_S = V_T = V$), Equation (6) can be simplified to (3):

$$g = \int_V \frac{B(\mu r)e^{-\mu r}}{r^2}\,dV \qquad \text{(Eq. 8a)}$$

where buildup is explicitly accounted for and is approximately given by values for the exposure buildup factor for an isotropic point source (4). Alternately, the geometry factor can be approximated as follows

$$g = \int_V \frac{e^{-\mu_a r}}{r^2}\,dV \qquad \text{(Eq. 8b)}$$

where the effect of buildup has been approximated by setting B equal to unity and replacing the linear attenuation coefficient μ by a linear energy absorption coefficient μ_a (5).

Equations 8(a) and 8(b) represent the geometry factor at a specific dose point. In many circumstances, an average geometry factor $\bar{g}$ is of interest. The latter can be calculated from the integral (3):

$$\bar{g} = \frac{1}{V}\int g_P\,dV \qquad \text{(Eq. 9)}$$

GEOMETRY FACTORS FOR A CYLINDER

Because of the difficulty in obtaining analytical solutions for a cylindrical geometry, Equations 8(a) & (b) were numerically solved to determine the geometry factors g_c at the center of a cylinder and g_P at any point in the cylinder. The latter were then integrated according to Equation (9) to estimate $\bar{g}$, the average value for a cylinder. A comparison of estimated geometry factor values with those in the literature indicated that data values in an often quoted literature source (6) were flawed. Further details can be found in Reference 2.

Sample calculations using PC-CAGE are summarised in Table I. These were performed for three cylinder sizes (Cylinders A, B and C) each assumed to be filled with water (properties of water are considered to approximate those of spent resin) and for a 1 MeV photon. Dimensions for Cylinder B correspond to the maximum radius and height limits for data published in the literature (6). Dimensions for Cylinder C correspond to OPG's spent resin container. For each case, three sets of geometry factors were computed:

- No buildup - calculations are based on Equation 8(a) where *B* was set equal to 1 and μ equaled 0.0706 cm^{-1} (7).
- Approximate buildup correction considered - calculations are based on Equation 8(b) with the corresponding value of μ_a equal to 0.0311 cm^{-1} (7).
- Explicit buildup correction considered – calculations are based on Equation 8(a) with μ equal to 0.0706 cm^{-1} (7) and a Taylor's form (7) for the buildup factor was used.

For each case, the calculated set of geometry factors consisted of g_c, the geometry factor at the center of the cylinder and $\bar{g}$, the average geometry factor for the entire cylinder. The sample calculations in Table I indicate the following:

- Neglecting buildup, in the case of large cylinders, can result in an error of greater than 100% (Column 7).
- Compared to the explicit treatment for the buildup factor, the approximate treatment (where B is set equal to unity and μ_a replaces μ) results in $\bar{g}$ being under-estimated by less than 25% (Column 8). Thus, the approximate treatment may suffice in many cases of practical interest. This treatment is computationally less intensive.
- The ratio $\bar{g}/g_c$ appears to be approximately constant indicating the feasibility of calculating $\bar{g}$ from the less computationally demanding calculation of g_c.

Table I. Estimated Geometry Factors for a Cylinder – Sample Calculations

	Radius (cm)	Height (cm)	Geometry Factor (cm)			Percent Difference	
			No Buildup Considered (I)	Buildup Considered		(III-I)/I × 100	(III-II)/II × 100
				Approx. (II)	Explicit (III)		
Cylinder A	10	10	g_c=80.5 $\bar{g}$=57.2 $\bar{g}/g_c$=0.71	g_c=94.9 $\bar{g}$=68.0 $\bar{g}/g_c$=0.72	g_c=111 $\bar{g}$=79.6 $\bar{g}/g_c$=0.72	38 39 -	17 17 -
Cylinder B	35	100	g_c=169 $\bar{g}$=130 $\bar{g}/g_c$=0.77	g_c=297 $\bar{g}$=217 $\bar{g}/g_c$=0.73	g_c=370 $\bar{g}$=267 $\bar{g}/g_c$=0.72	119 105 -	25 23 -
Cylinder C (OPG's spent resin container)	81.5*	170*	g_c=178 $\bar{g}$=143 $\bar{g}/g_c$=0.80	g_c=381 $\bar{g}$=286 $\bar{g}/g_c$=0.75	g_c=435 $\bar{g}$=337 $\bar{g}/g_c$=0.78	144 136 -	14 18 -

*Overall dimensions

APPLICATION TO ONTARIO POWER GENERATION'S SPENT IX RESIN

Radiolytic Gas Generation

Equation (9) was used to estimate the potential for H_2 gas generation in OPG's spent, PHT and moderator resin containers. The geometry factors were determined for a waste cylinder of height 146 cm and radius 80.865 cm, which dimensions are consistent with a waste volume of 3 m^3. For simplicity, the point geometry factors were based on the buildup treatment according to Equation 8(b). The value of the average geometry factor was substituted in Equation (7) to obtain the average dose rate in keV/m^3/s; this was multiplied by the container volume to obtain the average dose rate in keV/s.

For the principal gamma emitters present in the waste, results obtained using Equation (9) are compared in Table II with estimates obtained assuming complete deposition of the gamma decay energy within the package. Results based on the two approaches (for individual nuclides and their totals) are surprisingly comparable for each type of resin container. Instead of using Equation 8(b), the explicit consideration of buildup (Equation 8(a)) will increase the estimate based on geometry factors by approximately 18% (see Table I). Taking this into account, the results indicate that about 80% of the gamma energy originating in both types of resin containers is deposited internally; of the balance, a small fraction is deposited within the container wall material (see next sub-section) and the rest escapes the container wall and is thus responsible for the observed radiation fields.

To support the above conclusion, it was of interest to estimate the order of magnitude of the external radiation field that would result from escape of 20% of the source gamma energy. The case of the PHT resin container with a total source gamma emission energy of ~ 5.4 10^{15} keV/s (includes contributions from radionuclides besides those listed in Table II) was considered for this purpose:

> A sphere, equivalent to the 3 m^3 volume of the resin container would have a radius of 0.89 m. Because radiation fields are typically reported at 30 cm (1 ft) from the surface, the surface area for estimating the flux would correspond to a sphere with radius 0.89 + 0.30 m and equaled 17.9 m^2. Assuming each escaping photon has an energy of 1.25 MeV (average energy of Co-60 photon emission), the flux at 30 cm from the surface of the sphere is 20% x 5.4 10^{15}/ (1.25 x 1000 x 17.9 x 100) or 4.8 x 10^6 photons/s/cm^2. Based on a conversion factor of 1.98 x 10^{-6} R/h / photons/s/cm^2 (1), the escaping flux will give rise to a radiation field of ~9.6 R/h.

The order of magnitude of the estimated radiation field is consistent with observed fields for PHT resin containers and also with estimates obtained using Microshield. Thus, the observed or estimated radiation fields are in keeping with the escape of ~20% of the source gamma energy or alternately with ~ 80% of the source gamma energy being deposited within the resin waste.

Table II. Rate of Energy Deposition Within OPG's Spent Resin Containers - Comparison Between Estimates Based on Geometry Factors and Those Based on the Complete Deposition of Gamma Source Energy

Principal Gamma Emitters	Activity[a] (Ci/m^3)	Average Emission Energy, E (keV)	Total Intensity, I (%)	Photon Flux, S_V ($\gamma/s/m^3$)	Linear Energy Absorption Coeff.[b], μ_a (m^{-1})	Average Geometry Factor, $\bar{g}$ (m)	Rate of Energy Deposition (keV/s)	
							Based on Geometry Factor	Assuming Complete Deposition
Primary Heat Transport Resins								
Cs-137	1.60E+01	608.1	98.3	5.82E+11	3.29	2.63	7.31E+14	1.06E+15
Cs-134	6.30E+00	695.1	223.7	5.21E+11	3.25	2.66	7.48E+14[c]	1.09E+15
La-140	1.20E+01	1043.5	213.6	9.48E+11	3.09	2.76	2.01E+15[d]	2.97E+15
						Total	3.49E+15	5.12E+15
Moderator Resins								
Tb-160	4.60E-01	608.3	177.8	3.03E+10	3.29	2.63	3.81E+13	5.52E+13
Gd-153	7.40E+00	53.3	207.3	5.68E+11	4.18	2.18	6.59E+13	9.08E+13
Co-60	2.10E+00	1252.9	200	1.55E+11	2.98	2.84	3.92E+14	5.84E+14
						Total	4.96E+14	7.30E+14

a Based on Evans and Husain (8)
b From Lamarsh (7) and Jaeger et al (5)
c A value of 7.57 E+14 keV/s was estimated considering the detailed emission energies for Cs-134 instead of the average emission energy.
d A value of 2.48 E+15 keV/s was estimated considering the detailed emission energies for La-140 instead of the average emission energy.

Radiation Exposure of Container Material

While the average geometry factors are pertinent for the calculation of the radiolytic gas generation rates, the point geometry factors at the top, bottom and sides of the cylinder (their values are lower than that of the average geometry factor) are relevant for the estimation of dose to the container material. Such estimates would be of interest when considering alternate materials of construction for the shipping container. Note that the accurate estimation of surface geometry factors or surface dose rates is generally beyond the capability of shielding codes such as Microshield.

For the average Co-60 emission energy of 1.25 MeV (μ_a = 0.0298 cm^{-1}), Figures 2 (a) & (b) illustrate the variation of the point geometry factors as a function of the radial and axial distances from the base of the 3 m^3 resin container (radius 80.9 cm and resin bed height of 146 cm). As shown in Figure 2(a), the geometry factor at the ends decreases from a value of 2 m at the axis to a value of 0.854 m at the periphery. Values at the sides of the cylindrical resin volume, as expected, are symmetrical about the mid-plane and range from a high value of 1.67 m at the mid-plane to 0.854 m at the top or bottom end of the resin bed. The geometry factor values at the ends and sides are significantly lower than the values for the central (3.91 m) and average (2.89 m) geometry factors.

The end and side point geometry factors estimated according to Equations 8 (a) & (b) strictly apply to the exterior surface of the cylindrical resin volume but may be assumed to also apply approximately to the container material surrounding the resin. This is equivalent to considering Equation (8) as being a reasonable approximation of Equation (6) when $V_s \neq V_T$. This assumption is likely to be conservative for a cylindrical package geometry. To estimate the absorbed dose rate in the container material, Equation (7), after conversion from a volume to a mass basis, should, however, be based on the μ_a/ρ value for the container material rather than that for resin; in reality, at most energies of interest, this distinction is of no practical significance (7).

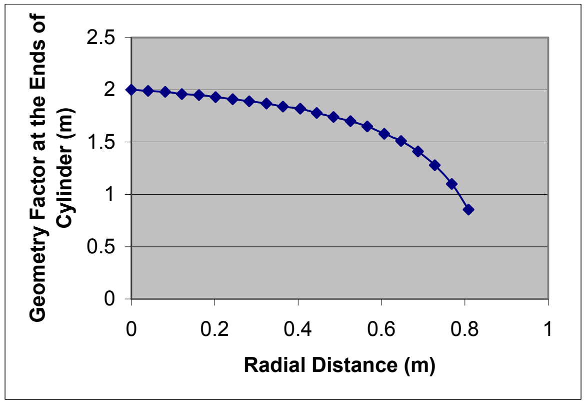
Based on Figures 2 (a) & (b), the container material would experience the maximum dose at the center of its bottom end (note that the top end of the resin container will be separated from the top of the resin bed by the headspace and will, therefore experience a somewhat lower dose rate) where *g* has a value of 2 m. A calculation for the estimation of the absorbed dose rate to the container material is illustrated below based on 1 Ci/m^3 Co-60 (average photon emission energy of 1.25 MeV and 2 photons per disintegration). The source photon flux,S_v corresponding to 1Ci/m^3 Co-60 equals 3.7 x 10^{10} x 2 or 7.4 10^{10} γ/s/m^3. Rewriting Equation (7) as

$$\dot{D}_\gamma = \frac{S_V}{4\pi} E \frac{\mu_a}{\rho} g \qquad \text{(Eq. 7a)}$$

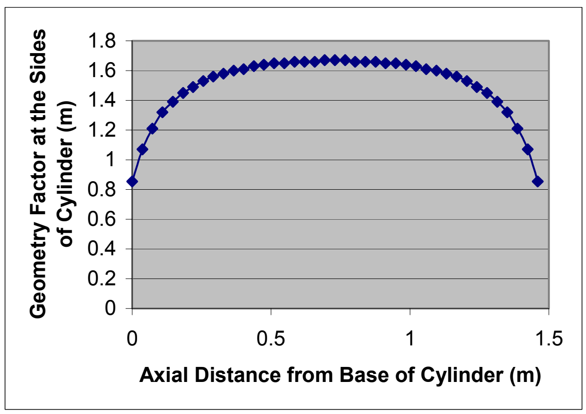
where the absorbed dose rate now has units of MeV/(s kg) and using the factor 1.6x10^{-11} to convert MeV/kg into rads, yields the following estimate for the absorbed dose rate in the container material:

Dose rate (rads/h) = 7.4 x 10^{10} x 1.25 x μ_a/ρ x $g/(4\pi)$ x 1.6 x 10^{-11} x 3600 = 424 μ_a/ρ x g

Substituting the values of 2.98 x 10^{-3} m^2/kg for μ_a/ρ and 2 m for *g*, yields an absorbed dose rate estimate of ~2.5 rads/h. Applying the 18% correction for the explicit treatment of buildup, yields a dose rate estimate of ~3 rads/h. Because the value of μ_a/ρ in the energy range 0.8-1.5 MeV is relatively constant for various materials (7), the absorbed dose estimate will be insensitive to the type of container material.



(a)



(b)

Fig. 2. Variation of Point Geometry Factor at a) Ends and b) Sides of 3 m^3 Resin Container Estimated for the Average Co-60 Photon Emission Energy of 1.25 MeV.

SUMMARY AND CONCLUSIONS

Estimation of the contribution from gamma emitters to the radiolytic gas generation rate in a waste package requires the computation of an average geometry factor.

Hitherto, no satisfactory method existed for estimating the geometry factors for a cylindrical package. This deficiency was recently addressed by formulating an expression for the point geometry factor and further integrating it over the volume of the cylinder. The formulation developed takes into account the effect of photon buildup arising from scattering events within the cylinder. Because of the complexity of the formulation, a prototype code called PC-CAGE was used to numerically solve the integrals involved. Based on any supplied dimensions for a cylinder and a value for either the linear energy absorption coefficient or the linear total attenuation coefficient (the coefficients correspond to the photon emission energy of interest and the material in the cylinder), the code outputs values for the central, surface (including top and bottom ends of cylinder) and the average geometry factors.

Calculations of radiolytic gas generation rate for OPG's resin containers were performed using PC-CAGE and also by assuming that the energy associated with gamma decay was completely deposited within the resin matrix. Results based on the two approaches (for individual nuclides and their totals) were surprisingly comparable indicating that about 80% of the source gamma energy in containers filled with either PHT or moderator resin, is deposited within the resin bed. In general, the fraction of source gamma energy which is deposited within the matrix will depend on the dimensions of the cylinder, the material within the cylinder and the photon energy; the fraction deposited increases with dimension of the cylinder and decreases with photon energy.

Of the 20% of the source gamma energy that escapes the OPG resin matrix, a fraction is deposited within the container material and the balance is responsible for the radiation fields observed on the outside of the resin containers. Although a complete energy balance was outside the scope of the calculations presented here, the magnitude of the energy absorbed by the container material and the magnitude of the external radiation field were both assessed.

Corresponding to 20% escape of the source energy and approximating the cylindrical container with a sphere of equivalent volume, the radiation fields @ 30 cm from the container surface was estimated to be ~ 10 R/h. This estimate is consistent with the magnitude of observed radiation fields for OPG containers containing PHT and moderator resins.

PC-CAGE was used to estimate the dose received by the material of the container. Calculations for OPG's 3 m^3 container indicated that the maximum value of the point geometry factor occurs at the centre of the top and bottom end surfaces of the source cylinder. Because the headspace in the container separates the top of the resin container from the top of the resin bed, the container material will experience the maximum absorbed dose rate at the center of its bottom end. Based on a Co-60 activity of 1 Ci/m^3, the estimated maximum absorbed dose rate experienced by the OPG container material was estimated to be ~ 3 rads/h.

ACKNOWLEDGEMENTS

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