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ScienceDirect
Journal of Nuclear Materials 359 (2006) 1–7
journal of nuclear materials
www.elsevier.com/locate/jnucmat

Geometrical α- and β-dose distributions and production rates of radiolysis products in water in contact with spent nuclear fuel

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Received 27 December 2005

Abstract

A mathematical model for the dose distribution and production rates of radiolysis products in water surrounding spent nuclear fuel has been developed, based on the geometrical and energetic properties of radiation. The nuclear fuel particle is divided into layers, from which the radiation emits. The water is likewise divided into layers, where the doses are distributed. The doses are stored in vectors which are added to determine the total dose rate. A complete inventory with over 200 radionuclides has been used as input data for the model. The purpose of the model is to describe the geometrical dose distribution as a function of fuel age and burn-up, to be used as input data for kinetic modeling of the fuel dissolution. The results show that the β-dose contribution close to the spent fuel surface is negligible. Also, the variation in the relative α/β dose contribution between different ages and burn-ups is insignificant. The α- and β-dose rates vary between different burn-ups of the same age; the younger the fuel is, the larger is the difference. Exponential functions have been fitted to the relations between fuel age and average dose rate, giving useful expressions for determining average dose rates for fuel ages other than those covered in this work.

1. Introduction

The environmental impact of nuclear power is an issue being continuously debated [1]. One of the key issues is the handling of the inevitable by-product, i.e., the highly radioactive spent nuclear fuel. Several countries plan to build deep repositories containing multiple natural and artificial barriers for spent nuclear fuel in order to protect the biosphere from contamination until the radioactivity of the fuel has declined to harmless levels. The safety assessment of such a repository relies on detailed knowledge about plausible processes for the dissolution of spent nuclear fuel under repository conditions [2–4], transportation and retention of radionuclides in the biosphere over time periods exceeding reasonable times for experiments by many orders of magnitude. Hence, very reliable models allowing fairly extreme extrapolations are necessary for this purpose.

As groundwater in deep repositories is expected to be reducing and since the fuel matrix, UO_2, has very low solubility under these conditions, the only possible pathway for release of radionuclides is by

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doi:10.1016/j.jnucmat.2006.08.001

radiolytical formation of oxidants causing dissolution of the fuel matrix [5]. Modeling of this process requires detailed knowledge about the geometrical dose distribution in the groundwater surrounding the spent fuel, the rate constants for all processes causing oxidation of the fuel matrix, i.e., UO_2, and the rate constants for dissolution of the fuel matrix as well as of the individual radionuclides. A model based on these entities constitutes the source term for a performance assessment of a spent nuclear fuel repository.

The very basis for such a source term is the geometrical dose distribution, i.e., the radiation dose rate as a function of distance from the fuel surface [6,7]. Furthermore, it is of vital importance to distinguish between the dose rates of α-, β- and γ-radiation since the yield of radiolysis products, the *G*-value, differs significantly between α-radiation on one hand and β- and γ-radiation on the other hand. Previous works have shown that empirical equations can be used to successfully describe the relation between energy and range for α-particles [7]. In this work, we have developed a model describing the geometrical dose distribution of α- and β-radiation from spent fuel and the resulting production rates of radiolysis products in water based on the spent fuel radionuclide inventory.

2. Methods

2.1. α-radiation

The concept of the model is to determine the geometrical dose rate distribution, and hence the primary radiolysis production rates in water surrounding a spent nuclear fuel surface, by modeling each of the steps necessary:

- The production rates of radiolysis products are determined by the dose rate in water, and the *G*-values.
- The dose rate in water is determined by the specific activity [8], decay energy [9], and the distance from the fuel surface.

The specific activities and decay energies are achieved from the composition of the spent nuclear fuel. The radiation energy determines its range in water and the fuel matrix. The range in uranium dioxide defines the active volume, i.e., the part of the spent fuel body that is able to contribute energy to the surrounding water body. The range in water gives the volume in which the energy is distributed. Also, the geometrical distribution of the radiation is accounted for, i.e., the isotropic nature of radioactive decays. These calculations will be made for each of the (up to 200) radionuclides occurring in spent nuclear fuel.

2.2. Activity calculations

The dose rate is determined by activity and particle energy. In order to calculate the dose rate in the water volume, the number of particles leaving the fuel surface, and their energy, must be determined. The maximum range an α-particle with energy E_0 can travel in water and uranium dioxide is calculated, using the empirical equations [7]:

$$\delta_w = 0.776E_0^2 + 2.738E_0 + 0.759, \quad (1)$$

$$\delta_{UO_2} = 1.118E_0^{3/2}. \quad (2)$$

Since the distance an α-particle can travel is small compared to the radius of the emitting body, the fuel surface is assumed to be planar. The volume of the part of the active material from which radioactive decay may contribute is calculated from δ_{UO_2}. Effective activity, contrary to specific activity, concerns the activity that affects the surroundings. Two factors affect the effective activity:

- Range
- Geometry

The range factor considers the fact that α- and β-particles emitted deep enough in the fuel are unable to reach the surface. The geometrical factor considers the isotropic nature of radioactive decay, i.e., that an emitted particle can have any direction. Although the path is considered linear, most particles will not travel perpendicular to the surface, and many will travel in opposite direction from the surface. A third factor, self-screening, affects the final dose rate. This will reduce the energy of the emitted particle.

The volume of the part of the spent fuel particle that is able to emit radiation to the surface is calculated by multiplying the surface area with the maximum range of the radiation in the material, thus assuming the range is considerably smaller than the spent fuel pellet. The total activity is achieved by multiplying the active volume with density, and activity per mass, which commonly serves as input data for spent fuel.

At maximum depth, a particle must travel perpendicular to the surface to be able to reach out

of the fuel particle, resulting in a very small fraction of the particles emitted from that depth reaching the surface. The fraction of particles escaping to the surface is determined by the depth. The particles emitted from a certain point source at a certain depth exit at the surface in a circle shaped area with center above the point where the particles are emitted. The radius of the circle at the surface is:

$$r = \sqrt{\delta_{UO_2}^2 - y^2}. \quad (3)$$

The activity contribution from each depth can be calculated and the sum of contributions from all depths between δ_{UO_2} and 0 gives the total flux of α-particles entering the surrounding water. The number of depths, which determine the resolution of the calculations, are set as a fixed value. When determining the fraction of activity that is emitted from the fuel surface, the area of a spherical segment is compared to the area of a sphere. This is shown in Fig. 1.

Area of spherical segment with center at depth y:

$$A_{ss} = 2\pi\delta_{UO_2}(\delta_{UO_2} - y). \quad (4)$$

Area of sphere with center at depth y:

$$A_s = 4\pi\delta_{UO_2}^2. \quad (5)$$

Fraction escaping from layer at depth y:

$$\frac{A_{ss}}{A_s} = \frac{2\pi\delta_{UO_2}(\delta_{UO_2} - y)}{4\pi\delta_{UO_2}^2} = \frac{(\delta_{UO_2} - y)}{2\delta_{UO_2}}. \quad (6)$$

Hence, at maximum depth ($y = \delta_{UO_2}$), no α-particles are emitted from the fuel surface, while at minimum depth ($y = 0$) 50% of the α-particles are emitted.

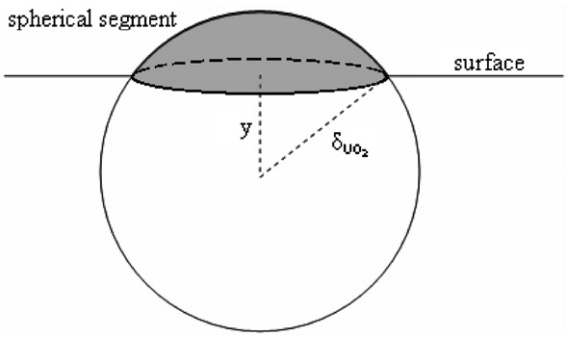

Fig. 1. The surface area of the spherical segment (shaded) corresponding to the number of α-particles emitted from the fuel surface, compared to the surface area of the sphere, to determine fraction of activity escaping.

The trapezoid method is used to numerically integrate the fraction of activity escaping from a certain depth, from the surface to the maximum depth. The fraction of activity escaping the fuel is shown as a function of depth in Fig. 2.

2.3. Energy calculations

Particles emitted from a certain point will reach the surface in a circular area. More particles will exit on the rim of the circle with low energies, while fewer will exit close to the center of the circle with high energies. The radius is the maximum range in the fuel matrix, δ_{UO_2}, while h is the height of a spherical segment, i.e., the difference between δ_{UO_2} and the depth, y. The effective range in water δ_{eff}, is the distance a particle travels perpendicular to the spent fuel surface, as shown in Fig. 3. The energy distribution of particles from a certain depth, y, is achieved by integrating numerically over the spherical segment:

$$E_y = \int_0^{y-\delta_{UO_2}} 2\pi\delta_{UO_2} h E(h)\,dh. \quad (7)$$

The equation is changed into a function of angle:

$$E_y = \int_0^{\pi} 2\pi\delta_{UO_2}(\cos\alpha \cdot \delta_{UO_2} - y)E(\alpha)\,d\alpha, \quad (8)$$

where $E(\alpha)$ is the energy of a particle when leaving the surface at the angle α. The energy of a particle as a function of the distance it has traveled in uranium dioxide after being released can be derived from Eq. (2):

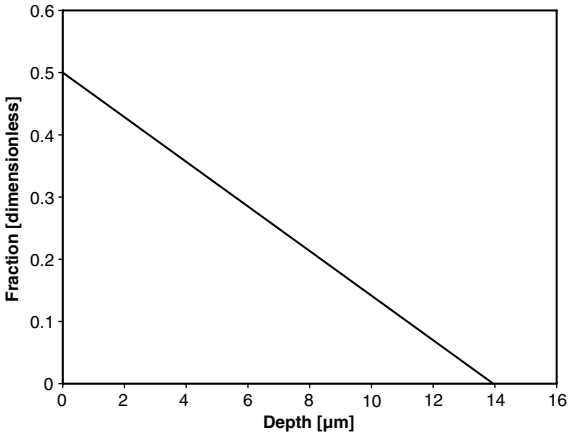

Fig. 2. The fraction of activity escaping the surface as a function of depth.

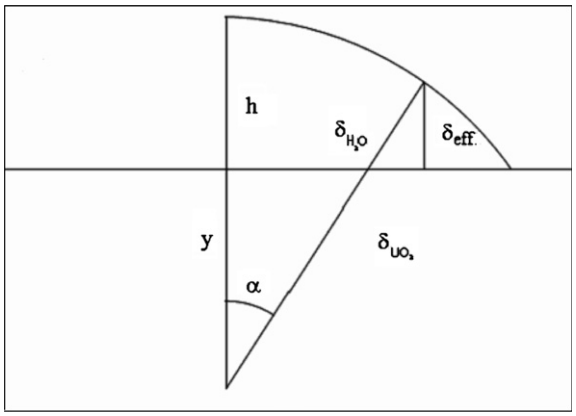

Fig. 3. Definition of effective range in water.

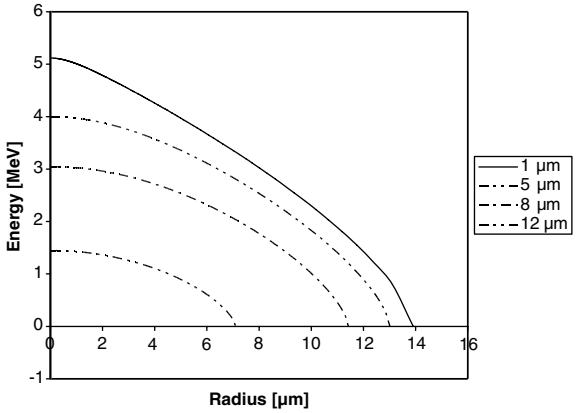

Fig. 4. Energy as a function of distance from the center of origin for α-particles released from different depths.

$$E(\delta) = \left(\frac{\delta_{UO_2} - \delta}{1.118}\right)^{3/2}. \quad (9)$$

The distance traveled as a function of the release angle:

$$\delta(\alpha) = \frac{y}{\cos\alpha}. \quad (10)$$

Combining Eqs. (9) and (10) gives the energy of a particle as a function of the angle:

$$E(\alpha) = \left(\frac{\delta_{UO_2} - \frac{y}{\cos\alpha}}{1.118}\right)^{3/2}. \quad (11)$$

The energy distribution of particles emitted from a certain depth, y, as a function of angle, derived from Eqs. (8) and (11):

$$E_y = \int_0^{\pi} 2\pi\delta_{UO_2}(\cos\alpha \cdot \delta_{UO_2} - y) \cdot \left(\frac{\delta_{UO_2} - \frac{y}{\cos\alpha}}{1.118}\right)^{3/2} d\alpha. \quad (12)$$

The energy as a function of distance from the point on the surface right above the point where an α-particle is released is shown in Fig. 4.

The energy distribution of the α-particles is obtained from the relation between particle energy and the fraction of activity escaping the surface. The distribution is calculated for each depth and added together, as can be seen in Fig. 5.

The energy flux at the surface is calculated from the mean surface energy for α-particles originating from a certain depth multiplied by the effective activity. The mean energy is calculated for each depth, and weighted using the relative activity for that depth.

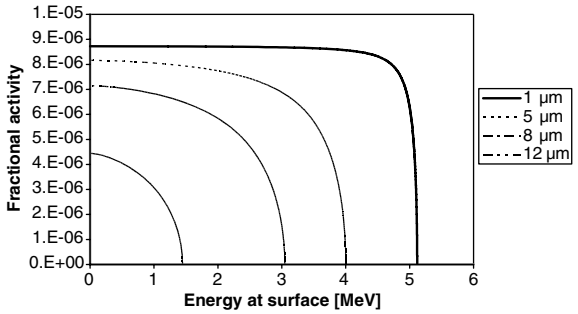

Fig. 5. Relative energy distribution of released α-particles from different depths. From top to bottom: 1, 5, 8 and 12 μm.

2.4. Dose distribution calculations

When a particle released inside the active material reaches the surface, the particle's energy and angle to the surface gives the effective range. The particle energy gives the range in water; the effective range is the distance the particle travels perpendicular to the surface.

$$\delta_{eff} = y\frac{\delta_w}{\delta_{UO_2}}. \quad (13)$$

The energy of each particle is assumed to be distributed uniformly in the water volume consisting of the layer closest to the fuel surface limited by the effective range. The water body is divided into thin layers.

For each depth, and each angle, the energy distributed as a function of distance from the surface is stored in vectors. Each element corresponds to a dose rate per mass at a certain distance from the surface. To get the total dose rate, all dose contributions, i.e., the vectors, are added together.

2.5. β-radiation

The method is identical to the method used as for α-radiation, with minor differences. Each β-emitter must be handled separately, because of the large variation (0.007–0.958 MeV) in energies.

For α-radiation, a function exists that relate range to energy. The corresponding relation for β-radiation is a polynomial that contains a constant term, which means that the range is not zero for particles with no energy. To obtain a more realistic function between energy and range, the least squares method is used to determine the range of β-radiation in different materials as a non-polynomial. The relation between energy and range in lead is described by a function of the type [10]:

$$\delta = \frac{kE^m}{\rho}, \tag{14}$$

δ is the β-range in a material with density ρ. k and m are constants that are calculated from data for different materials. Ranges in four different materials for seven different energies are used [11] in determining the factors for the polynomial. The constants have been calculated to:

$$k = 4.4574,$$
$$m = 1.1127,$$
$$\delta = \frac{4.4574E^{1.1127}}{\rho}. \tag{15}$$

The resulting function is shown in Fig. 6.

2.6. Dose rate approximation

An approximation of the dose rate, D', in the surrounding of an α- and β-emitting particle can be done analytically with simple means. Dose rate is defined as the energy absorbed in a certain mass per time unit. The absorbing mass, in this case, is the body of water surrounding the emitting entity, as thick as the maximum range of each particles in water. The dose rate for each nuclide is given by:

$$D' = \frac{AE_\alpha}{m_{H_2O}}. \tag{16}$$

The dose rate approximation is made by calculating the mean dose rate for the dominating α- and β-emitting nuclides for the different spent fuel compositions. The total dose rate close to the surface (α-range) is the sum of all mean dose rates.

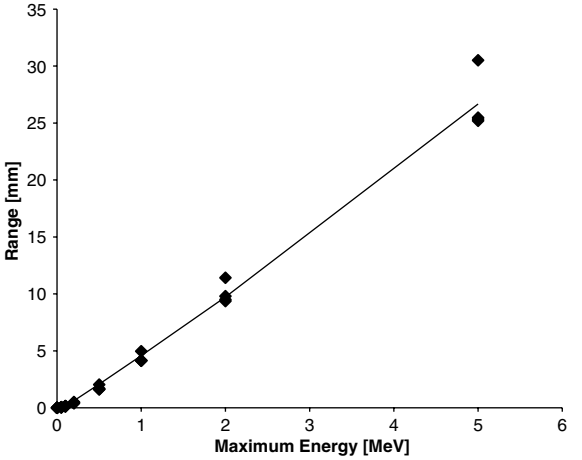

Fig. 6. β-range as a function of maximum β-energy for materials with density 0.7, 1.3, 2.6 and 11.3 g/cm^3.

The emitting and absorbing masses are defined as:

$$m_{fuel} = 4\pi r^2 \delta_{fuel} \rho_{fuel}, \tag{17a}$$
$$m_{H_2O} = 4\pi r^2 \delta_{H_2O} \rho_{H_2O}. \tag{17b}$$

The dose rate is then:

$$D' = \sum A_m E \frac{m_{fuel}}{m_{H_2O}} = \sum A_m E \frac{\rho_{UO_2} \delta_{UO_2}}{\rho_{H_2O} \delta_{H_2O}}. \tag{18}$$

The resulting dose rate is multiplied by factors accounting for geometry (0.25), and self-absorption (0.5). With the activity per mass attained from the decay calculations, the approximated value of the mean dose rate in the surrounding water close to the surface for spent fuel at 1000 years, 38 MWd/kg [8] is:

$$D' = 0.109 \text{ Gy/s}.$$

The complete model, including all nuclides in the inventory, gives the average dose rate close to the surface:

$$D' = 0.112 \text{ Gy/s}.$$

3. Results and discussion

The geometrical dose profile was calculated for 38 and 55 MWd/kg fuel burn-up at age 100, 1000, 10 000 and 100 000 years. In Fig. 7 the geometrical α-, β- and total dose profiles within the α-range for 100 year old 38 MWd/kg fuel is given.

As can be seen, the dose rate decreases dramatically with distance from the surface. The direct consequence of this is that there will be a sharp inherent

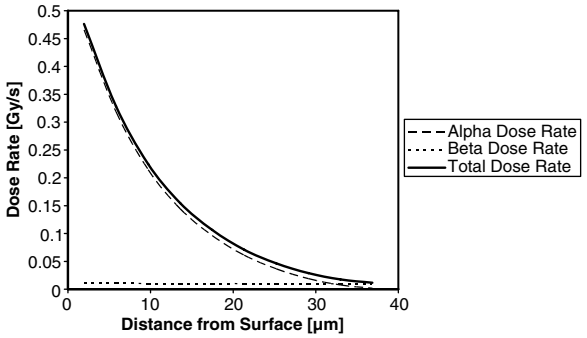

Fig. 7. Dose rate profile for the α-volume for 100 years old fuel with a burn-up of 38 MWd/kg.

concentration gradient for all radiolysis products. Hence, long-lived products will diffuse out from the surface into the bulk. Diffusion will counteract the relatively sharp dose profile. It should also be noted that the β-dose contribution to the total dose within the α-range is very small. This implies that only α-radiolysis need to be accounted for within the α-range.

The geometrical dose distributions for different burn-ups and fuel ages have the same profile. However, the absolute dose rates differ significantly. The average dose rates and the relative contributions from β-radiation for all the fuel ages and burn-ups studied are presented in Tables 1 and 2.

As can be seen, the average α-dose rate for the highest burn-up is 44% higher than for the lowest burn-up when comparing 100 years old fuel. For the oldest fuel (100 ky) the difference has been reduced to 31%. The difference between the two

Table 1
Average dose rates, and dose rate fractions for 38 MWd/kg fuel

	38 MWd/kg uranium			
	100 (y)	1000 (y)	10 (ky)	100 (ky)
Average α dose rate in α volume (Gy/s)	1.19E−01	2.87E−02	5.90E−03	5.44E−04
Average β dose rate in α volume (Gy/s)	9.54E−03	7.66E−05	2.65E−05	9.11E−06
Average β dose rate in β volume (Gy/s)	6.16E−04	9.25E−06	3.55E−06	1.17E−06
Average dose rate in β volume (Gy/s)	1.65E−03	2.53E−04	5.91E−05	5.28E−07
α dose fraction in α volume	0.926	0.997	0.996	0.984
β dose fraction in α volume	0.074	0.003	0.004	0.016

Table 2
Average dose rates, and dose rate fractions for 55 MWd/kg fuel

	55 MWd/kg uranium			
	100 (y)	1000 (y)	10 (ky)	100 (ky)
Average α dose rate in α volume (Gy/s)	1.71E−01	3.11E−02	6.06E−03	7.11E−04
Average β dose rate in α volume (Gy/s)	1.29E−02	1.56E−04	7.08E−05	1.37E−05
Average β dose rate in β volume (Gy/s)	8.38E−04	1.88E−05	9.77E−06	1.85E−06
Average dose rate in β volume (Gy/s)	2.33E−03	2.82E−04	6.68E−05	8.86E−06
α dose fraction in α volume	0.930	0.995	0.988	0.981
β dose fraction in α volume	0.070	0.005	0.012	0.019

burn-ups will also be reflected in the production of radiolytical oxidants as well as in the rate of fuel dissolution. It is also obvious that the average dose rate decreases by almost one order of magnitude as the fuel age increases by a factor 10. This is illustrated in Fig. 8.

As can be seen, we have used exponential functions to fit the estimated average dose rates as a function of time. The resulting Eqs. (19) and (20) can be used to estimate the average dose rates for fuel with burn-ups of 38 and 55 MWd/kg.

$$D' = 0.8653\mathrm{e}^{-1.775t}, \tag{19}$$

$$D' = 1.1321\mathrm{e}^{-1.8087t}. \tag{20}$$

3.1. Radiolysis calculations

Given the dose profile, the production rate for radiolysis products can be calculated as a function

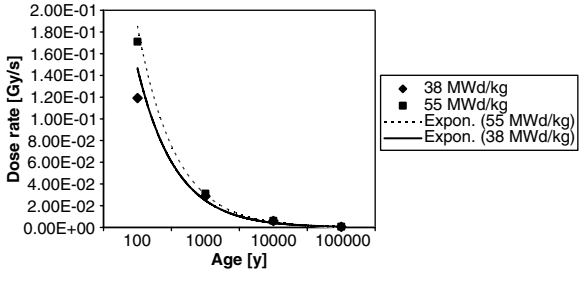

Fig. 8. Average α-dose rates for different burn-ups as a function of time.

Table 3
Production rates of radiolysis products in the α-volume for 1000 years old fuel at 38 MWd/kg

	Production rate (M/s)
e^-	1.84E−10
$H^\cdot$	6.26E−10
H_2	3.87E−09
H_2O_2	2.93E−09
HO_2	6.54E−10
$OH^\cdot$	7.20E−10

of distance from the surface by using the radiation chemical yield [12].

$$\frac{dC}{dT} = \left(G_\alpha D'_\alpha + G_\beta D'_\beta + G_\gamma D'_\gamma\right)\rho. \quad (21)$$

Using the average dose rate, the production rate of radiolysis products can be determined at a certain distance from the surface. The resulting production rates are given in Table 3.

4. Summary

A model describing the dose rate distribution and production of primary radiolysis products in the water surrounding a spent nuclear fuel pellet has been developed. The model uses data for all radionuclides present in spent fuel as input data, and accounts for geometric factors and loss of energy of emitted particles. The model describes the dose rate distribution and production rate of radiolysis products as a function of distance from the spent fuel surface, and the results agree with approximations based on the few radionuclides that contribute most to the dose rate.

Acknowledgements

The Swedish Nuclear Fuel and Waste Management Company (SKB) and the Swedish Nuclear Power Inspectorate (SKI) are gratefully acknowledged for financial support.

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