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Alpha, Beta and Gamma Dose Rates in Water in Contact with Used CANDU UO_2 Fuel

Débits de dose alpha, bêta et gamma dans l'eau en contact avec le combustible (UO_2) CANDU irradié

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ALPHA, BETA AND GAMMA DOSE RATES IN WATER IN CONTACT WITH USED CANDU UO_2 FUEL

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ABSTRACT

Alpha, beta and gamma dose rates in water, in contact with the reference used fuel in the Canadian Nuclear Fuel Waste Management Program, i.e., Bruce "A" CANDU fuel, burnup 685 GJ/kg U, are calculated as a function of cooling time. Procedures are described to calculate the dose rates for used CANDU fuels of different burnups from results obtained for the reference fuel. This information is needed to compare the results of leaching and corrosion experiments carried out with used CANDU fuels of different burnups and/or cooling times and to predict the effects of radiolysis of groundwater on used fuel oxidation and dissolution in a disposal vault.

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DÉBITS DE DOSE ALPHA, BÊTA ET GAMMA DANS L'EAU EN CONTACT AVEC LE COMBUSTIBLE (UO_2) CANDU IRRADIÉ

par

S. Sunder

RÉSUMÉ

Les débits de dose alpha, bêta et gamma dans l'eau en contact avec le combustible irradié de référence dans le cadre du Programme canadien de gestion des déchets de combustible nucléaire, c.-à-d. le combustible CANDU de Bruce «A», de combustion massique 685 GJ/kg d'U, sont calculés en fonction du temps de refroidissement. On décrit les méthodes qui permettent de calculer les débits de dose pour des combustibles CANDU irradiés de diverses combustions massiques d'après les résultats obtenus pour le combustible de référence. Ces données sont nécessaires à la comparaison des résultats des expériences de lixiviation et de corrosion effectuées avec des combustibles CANDU irradiés ayant des combustions massiques différentes et/ou ayant refroidi pendant des périodes différentes, et pour prévoir les effets de la radiolyse des eaux souterraines sur l'oxydation et la dissolution du combustible irradié dans une enceinte de stockage permanent.

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CONTENTS

1. INTRODUCTION

The concept of geological disposal of used nuclear fuel is being investigated in several countries [1-10]. The concept envisages sealing the used nuclear fuel in containers made of corrosion-resistant metal(s) and placing the containers in a disposal vault [1]. Transport by groundwater is the most likely mechanism for radionuclides to migrate from the used fuel bundles in the disposal vault to the biosphere. Therefore, an assessment of used-fuel disposal requires a prediction of the release rates of radionuclides from the fuel once contact with groundwater is established.

The great majority of radionuclides, >90%, are contained within the grains of the fuel pellets and will be released at a rate governed by the dissolution rate of the UO_2 fuel matrix. The problem of measuring and predicting UO_2 dissolution rates is complicated by the sensitivity of uranium solubility to redox conditions [10-12]. Although UO_2 is highly insoluble, under reducing conditions found in granitic groundwaters at the planned depth (~500 m to 1 km) of the disposal vault envisaged in the Canadian Nuclear Fuel Waste Management Program (CNFWMP) [1], the solubility of uranium increases by many orders of magnitude under oxidizing conditions. The redox conditions of the groundwater in contact with the used nuclear fuel may be modified due to the radiolysis of water by the ionizing radiation from the fuel.

The radiolysis of water produces both molecular and radical oxidants and reductants [13-18]. The concentration of the different species formed during radiolysis depends on both the nature of the ionizing radiation, and its dose rates in water. It is well known that low linear energy transfer (LET) radiation, e.g., beta and gamma, produces more radicals than high LET radiation, e.g., alpha. Alpha radiolysis of water results predominately in the formation of molecular radiolysis products. Therefore, it is necessary to know the dose rate in water for each of the different types of ionizing radiation associated with the used fuel in order to evaluate the effects of water radiolysis on the corrosion rate of the fuel.

The gamma dose rate in water, in contact with used CANDU® fuel, was reported recently for a fuel with a burnup of 685 GJ/kg U, the reference used fuel in the CNFWMP, as a function of cooling period [5]. The procedure to calculate gamma dose rate for other burnups is described here. Very little information is available on the dose rate in water due to alpha and beta radiation from used CANDU fuel. Garisto has calculated the dose rate in water due to alpha radiation for a limited burnup and cooling periods [19]. There is no report on the dose rate in water in contact with used CANDU fuel due to the beta radiation. Several reports give the alpha and beta activity in used CANDU fuel; however, they do not provide the dose rate in water [20,21]. Here we report dose rates in water due to alpha and beta radiation in water in contact with the reference CANDU fuel as a function of cooling time. Procedures to estimate the dose rates due to alpha, beta and gamma radiations for fuels of different burnups are also described.

2. DOSE RATE IN WATER DUE TO ALPHA AND BETA RADIATION

The procedure described here to calculate dose rates in water due to the alpha and beta radiation assumes that the radionuclides are uniformally distributed in the fuel. It disregards known variations in the concentrations of radionuclides in the used fuel [22]. Thus, the dose rates calculated are approximate average values. Also, the procedure is for water in direct contact with the UO_2 fuel, i.e., the cladding has been breached.

The dose rate in a thin target through which a beam of charged particles (alpha or beta) passes is given by the relation [14,23]:

$$D = K \cdot \phi \left(- \frac{1}{\rho} \cdot \frac{dE}{dx} \right) \tag{1}$$

where
D = dose rate ($Gy \cdot h^{-1}$),
K = a constant,
ϕ = particle flux ($cm^{-2} \cdot s^{-1}$),
ρ = density ($g \cdot cm^{-3}$),
$-dE/dx$ = linear stopping power ($MeV \cdot cm^{-1}$), and
$-(1/\rho)(dE/dx) = T$ = mass stopping power ($MeV \cdot cm^2 \cdot g^{-1}$).

Thus, the dose rate in the water layer in contact with the used fuel, due to alpha (beta) radiation, can be related to the dose rate in the fuel matrix (if the alpha (beta) flux density in the interface between the fuel and water is continuous) by the relation [14,23]:

$$\frac{D(H_2O)}{D(UO_2)} = \frac{\left[\frac{1}{\rho} \cdot \frac{dE}{dx} \right]_{H_2O}}{\left[\frac{1}{\rho} \cdot \frac{dE}{dx} \right]_{UO_2}} \tag{2}$$

Equation (2) is essentially the result of the boundary condition that the flux of the particle out from the fuel equals the flux into the water with the stoppage of particles right at the interface being neglected. Hence, one can obtain the dose rate in water, in contact with the fuel, if one knows the dose rate in the fuel and the ratio of the mass stopping power for the alpha (beta) particles of water and the fuel.

2.1 DOSE RATE IN WATER DUE TO ALPHA RADIATION

The mass stopping power for alpha particles is given by the Bethe-Block formula [13,23-25]:

$$T = -\frac{1}{\rho} \cdot \left[\frac{dE}{dx}\right] = k \cdot \frac{Z}{A} \cdot z^2 \cdot f(\beta,I) \qquad (3)$$

where k = a constant,
z = charge on the alpha particle,
Z = "atomic number",
A = "atomic mass", and
I = ionization potential, and
β = relativistic speed of the alpha particle.

Note, β is = V/C, where V = velocity of the alpha particles, and C = speed of light.

The function $f(\beta,I)$ is a weak function of I, i.e., it can be approximated by the same value. Thus, for two different media, the relationship becomes

$$\frac{T_a}{T_b} = \frac{\left[\frac{Z}{A}\right]_a}{\left[\frac{Z}{A}\right]_b} \qquad (4)$$

If a and b are H_2O and UO_2, respectively (i.e., $Z_a = 10$, $Z_b = 108$, $A_a = 18$ and $A_b = 270$), the value of this ratio is 1.39. The above value of the ratio of the stopping powers of water and UO_2 for alpha particles obtained from Equation (3) may be compared with the value obtained from Equation (1) using the literature values of the stopping powers for water and UO_2. Nitzki and Matzke [24] have measured the stopping power of UO_2 for the alpha particles. There are several references available for the stopping power of water for the alpha particles [13,14,25,26]. Table 1 compares the stopping powers of UO_2 and water for alpha particles of energy 5.3 MeV. Using the experimental value for the stopping power of water reported by Spinks and Woods [14] and the used fuel density of 10.4 g/cm^3 [21], one obtains a value of 1.38 for the ratio of the dose rates in water and UO_2 from Equation (2), a value in very good

agreement with that obtained using Equation (4). Note that Equations (2) and (4) give dose rates in water in small cracks inside the fuel. In order to obtain dose rates in water outside the fuel, but within the range of the alpha particles from the fuel, the dose rates calculated using these equations are further divided by 2 [19,23].

TABLE 1

LINEAR STOPPING POWER FOR 5.3-MeV ALPHA PARTICLES

Medium	-dE/dx keV/μm	Reference
UO_2	325	Nitzki and Matzke (1973)
H_2O	150	Swallow (1960)
H_2O	93	Allen (1961)
H_2O	43	Spinks and Woods (1990)[a]
H_2O	136	Spinks and Woods (1990)[b]

a Experimental value (page 65 of reference).
b Average literature value (page 17 of reference).

Figure 1 shows the alpha dose rate in water, in contact with used CANDU fuel with a burnup of 685 GJ/kg U (the reference used fuel in the CNFWMP), as a function of cooling time. The results shown in Figure 1 were calculated from the alpha dose rates in the fuel and Equation (4). Alpha dose rates in the fuel were calculated using the decay heat values reported by Smith et al. [20]. (Smith et al. give decay heat values in units of W/kg initial uranium. To obtain dose rate in the fuel it was assumed that 1 kg of CANDU fuel contains 0.88148 kg of initial uranium). The dose rates shown in Figure 1 are average values for a water layer outside the fuel and for thickness equal to or less than the range of the alpha particles in water, which is a function of their energy [14,19,27]. (Appendix A gives the ranges of the alpha-particles in water). It should be noted here that Garisto has shown that over 60% of the alpha-particle energy is deposited in the water layer of thickness less than 30% of the alpha-particle range (Figure 4 in Reference 19).

In order to obtain the alpha dose rate in water in contact with a fuel of burnup different than the reference fuel, we have plotted in Figure 2 the relative values of alpha dose rates as a function of burnup for different cooling periods (1, 10, 100, 1000 and 10 000 a respectively). These values were normalized to yield a value of 1 for a burnup of 685 GJ/kg U. Again, the

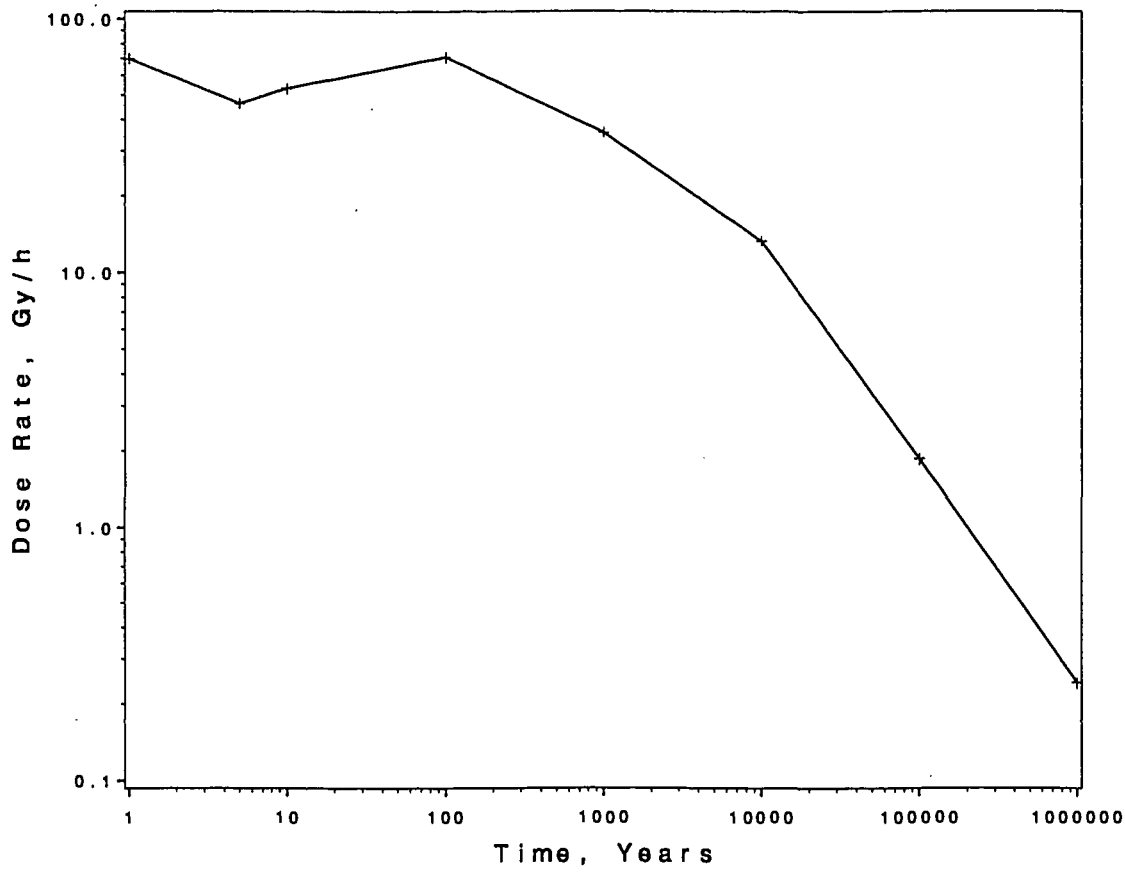

FIGURE 1: The Average Alpha Dose Rate in Water Layer of Thickness Equal to the Range of Alpha Particles (~ 35 μm), in Contact With Used CANDU Fuel, Burnup 685 $GJ \cdot kg^{-1}$ U, as a Function of Cooling Time.

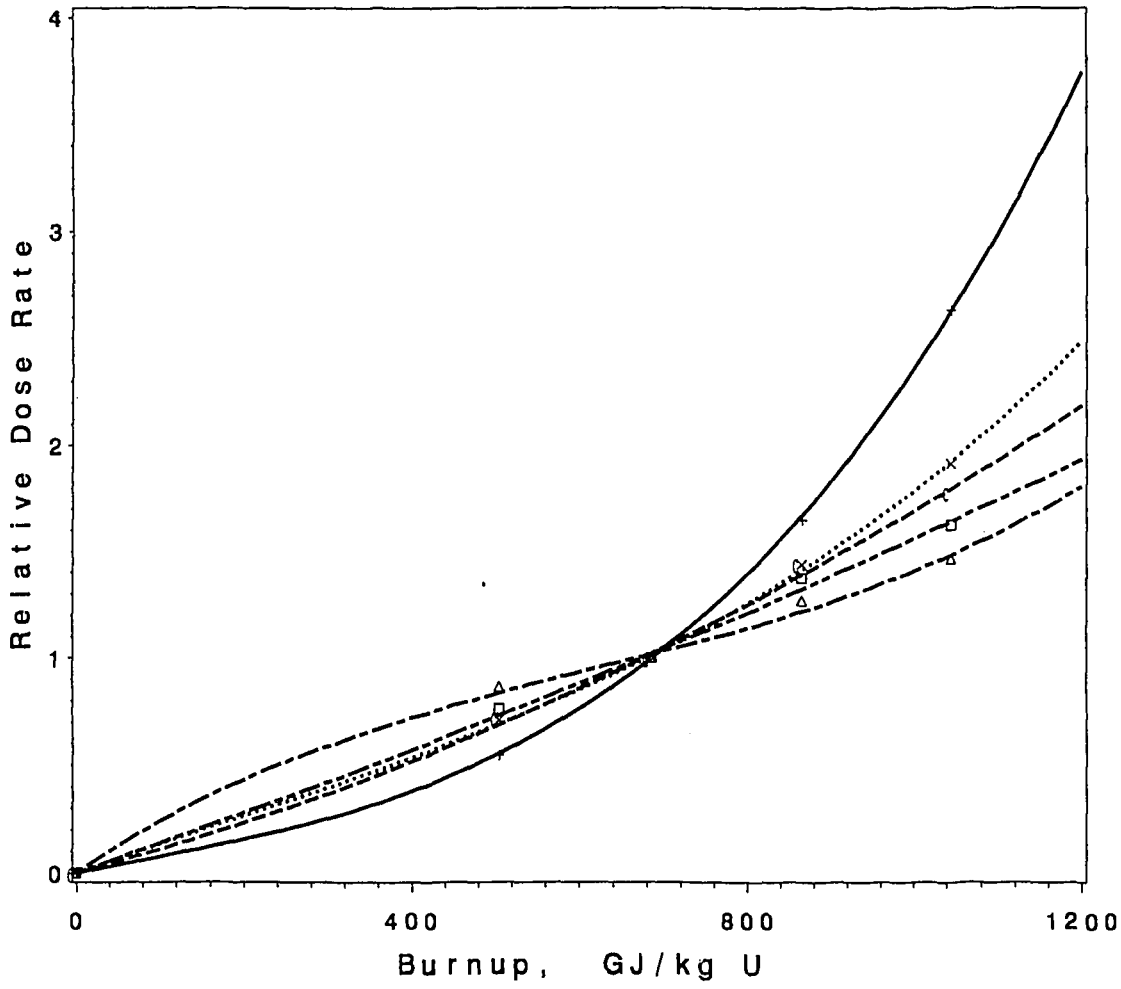

FIGURE 2: The Relative Alpha Dose Rate in Water Near the Surface of Used CANDU Fuel as a Function of Burnup after Different Cooling Periods: (a) 1 a, +; (b) 10 a, X; (c) 100 a, O; (d) 1000 a, □; and (e) 10 000 a, ∆.

decay heats reported by Smith et al. [20] were used to obtain the results shown in this figure. The relative values of the dose rate were fitted to a cubic equation. Despite the fact that alpha dose rates are not a linear function of burnup, one can use the plots shown in Figure 2 to estimate dose rates for different burnups and cooling times using the results given in Figure 1 for the reference fuel. One obtains the alpha dose rate for the fuel of a given burnup and cooling period by multiplying the relative value of the dose rate from Figure 2 by the dose rate for the reference fuel of the same cooling period. For example, to obtain the alpha dose rate for CANDU fuel with a cooling period of 1000 a and a burnup of 1045 GJ/kg U, one obtains the dose rate for the reference fuel from Figure 1 (35.3 $Gy \cdot h^{-1}$) and multiplies it by the relative value for the fuel of burnup 1045 GJ/kg U, from Figure 2 (1.625), Table 2. One can estimate the relative value of the alpha dose rate for cooling periods other than those used in Figure 2 by interpolating or extrapolating the results given in this figure.

TABLE 2

ALPHA DOSE RATES IN WATER IN CONTACT WITH USED CANDU FUELS STUDIED BY GARISTO

USED FUEL			DOSE RATE		
#	Burnup	Cooling Time	From Garisto's Data		This Work
			a	b	
	GJ/kg U	a	$Gy \cdot h^{-1}$	$Gy \cdot h^{-1}$	$Gy \cdot h^{-1}$
1	685	100	57.67	124.00	70.57
2	685	1000	27.33	58.76	35.29
3	685	10000	10.33	22.20	13.23
4	1045	1000	40.74	87.59	57.36

a Calculated from E_{out} values of Garisto, Appendix A, and using a water layer of thickness = 34.4 μm.

b Calculated from E_{out} values of Garisto, Appendix A, and using a water layer of thickness = 16.0 μm.

Table 2 compares the alpha dose rates calculated using the results shown in Figures 1 and 2 with those obtained from Garisto's results for the four fuels investigated by him. (The procedure followed to obtain alpha dose rates from Garisto's results is summarized in

Appendix A). Dose rates calculated from Garisto's results are slightly lower than those calculated in the present work, if one uses a water layer thickness, d, of 34.4 μm, i.e., the range of 5.2-MeV alpha particles, the maxima in the energy spectrum for CANDU fuel with a cooling period of 100 a [19]. On the other hand, the dose rates calculated using Garisto's results are higher than those calculated in the present work, if one uses a water layer thickness of 16 μm, i.e., the range of 2.93-MeV alpha particles, the average energy of the emitted alpha particles with an initial energy of 5.2 MeV according to Garisto's calculations [19].

2.2 DOSE RATE IN WATER DUE TO BETA RADIATION

The dose rate from beta particles in water in contact with the used CANDU fuel can be calculated using Equation (2) if one knows the beta dose rate in the fuel and the ratio of the stopping powers of water and the fuel (UO_2), as discussed above. The mass stopping power of water for beta particles, as a function of energy, is available in the literature [25]. However, there is no report on the stopping power of UO_2 for electrons. Therefore, we calculated the stopping power of UO_2 for electrons from the literature values of the stopping power of uranium and oxygen [25] using the additivity law described by Haissinsky [27]. According to this law, one can obtain the mass stopping power of a compound A_aB_b from the known stopping powers of A and B using the formula:

$$-\frac{1}{\rho}\frac{dE}{dx} = \frac{a\ A_A\left(-\frac{1}{\rho}\frac{dE}{dx}\right)_A + b\ A_B\left(-\frac{1}{\rho}\frac{dE}{dx}\right)_B}{M} \qquad (5)$$

where M is the molecular weight of compound A_aB_b and $A_A$and A_B are the atomic weights of the elements A and B, respectively.

The mass stopping powers for electrons in UO_2 calculated using Equation (4), are given in Table 3. This table also gives the ratio of the stopping powers in water and UO_2. The mass stopping powers decrease with the energy of the beta particles. However, the ratio of the mass stopping powers is a weak function of energy. The average energy of beta particles in the reference CANDU fuel with a cooling period of 10 a or more is about 0.22 MeV (as calculated from the data of Smith et al. [20]). Therefore, to calculate the dose rate in water, we have used a value of 2.0 for the ratio of the mass stopping power of water to that of UO_2 fuel for the beta particles in Equation (2). We used a procedure similar to that described in Section 2.1 for alpha radiation to calculate the dose rate in used fuel from the beta decay heat calculated by Smith et al. [20].

TABLE 3

MASS STOPPING POWER OF WATER AND UO_2 FOR BETA PARTICLES

Energy of Beta Particles MeV	Mass Stopping Power $MeV \cdot cm^2 \cdot g^{-1}$		Ratio of Stopping Powers
	H_2O[a]	UO_2	
0.01	22.570	9.35196	2.81949
0.05	6.607	3.24441	2.27123
0.10	4.120	2.12862	2.13251
0.20	2.798	1.51692	2.01006
0.40	2.152	1.22877	1.88772
0.60	1.965	1.16456	1.80607
1.00	1.857	1.16625	1.68818
2.00	1.857	1.29440	1.49758

a From ICRU Report 37 (Reference 25).

Figure 3 shows beta dose rates in water, calculated using Equation (2) and data of Smith et al. [20], as a function of cooling period near the surface of the reference CANDU fuel. Figure 4 show a plot of the relative values of the beta dose rates, obtained using the beta decay heat data [20], as a function of burnup for different cooling periods (1, 10, 100 and 1000 a). These values were normalized to yield a value of 1 for a burnup of 685 GJ/kg U. One can obtain the value of beta dose rates for burnup values other than 685 GJ/kg U using the data given in Figures 3 and 4 and following a procedure similar to that described for alpha dose rates (Section 2.1).

The dose rates shown in Figure 3 are average values for a water layer outside the fuel and of thickness equal to the range of the beta particles. It has been shown that for UO_2 corrosion studies only the dose rate in water layers less than 50 μm from the surface need be considered [28]. As the range of the beta particles is longer than 50 μm [14,27], one can use the dose rates obtained from Figures 3 and 4 for the used fuel corrosion studies. The variations in the beta dose rate in water layers at different distances from the beta particle source have been discussed by several workers [13,14,23,25,27,29]. They follow a similar distribution profile, as that observed for the alpha particles [19], if the distance from the source (fuel) is expressed in the dimensionless units of the fraction of the particle range in water (e.g., see Figure 8 in Reference 29). As the range of the beta particles is much longer than that of the alpha particles [14,27], the variation in the beta dose rate, in a water layer of thickness ~50 μm, is much smaller than the variation in the alpha dose rate.

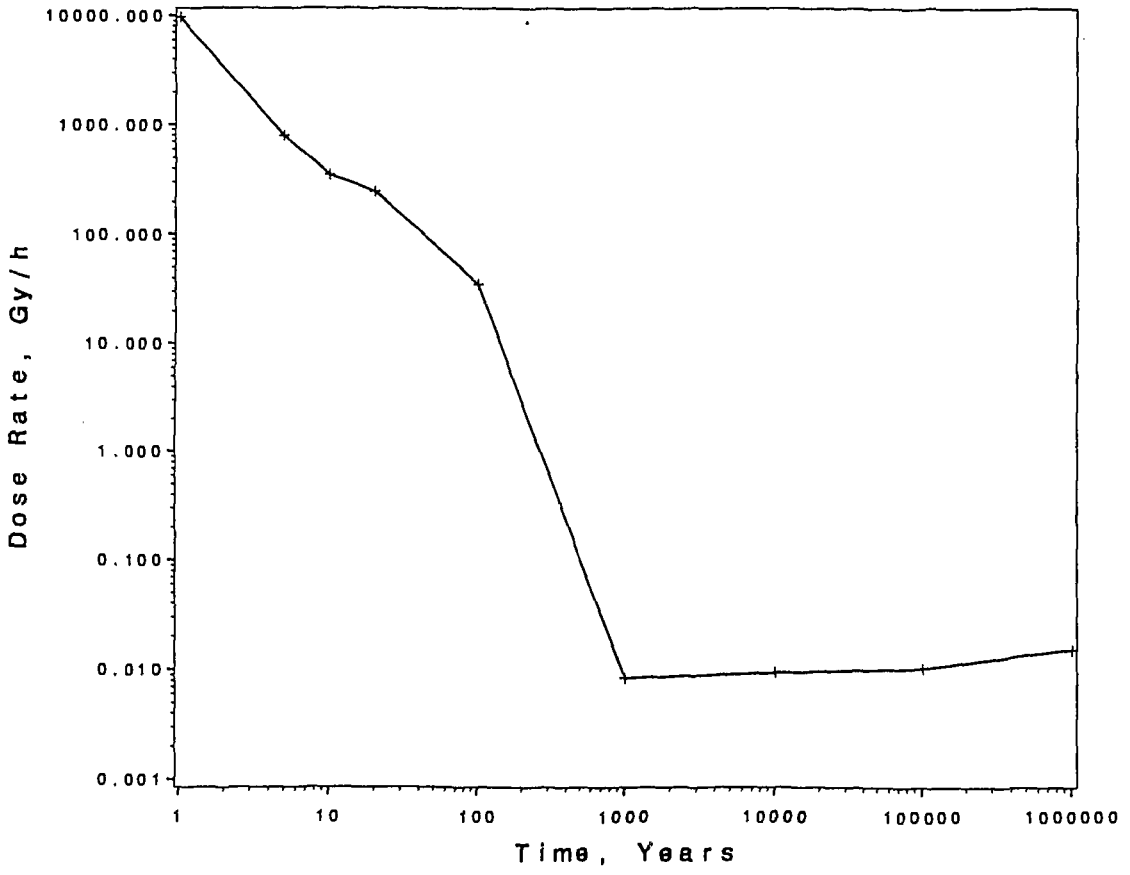

FIGURE 3: The Average Beta Dose Rate in Water Layer of Thickness Equal to the Range of the Beta Particles (~ 400 μm), in Contact With Used CANDU Fuel, Burnup 685 $GJ \cdot kg^{-1}$ U, as a Function of Cooling Time.

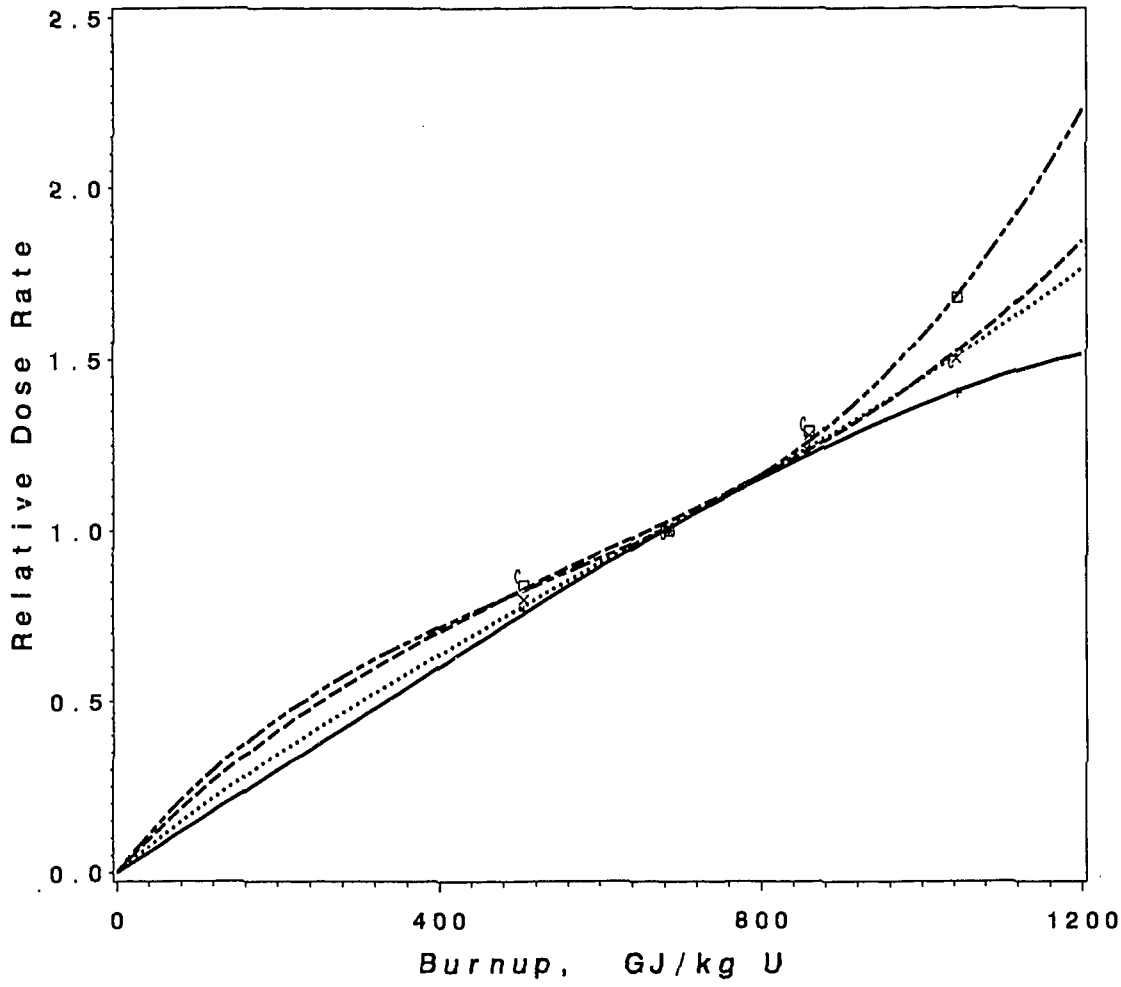

FIGURE 4: The Relative Beta Dose Rate in Water Near the Surface of Used CANDU Fuel as a Function of Burnup after Different Cooling Periods: (a) 1 a, +; (b) 10 a, X; (c) 100 a, O; and (d) 1000 a, □.

3. DOSE RATE IN WATER DUE TO GAMMA RADIATION

Figure 5 shows the gamma dose rate in contact with a bundle of the reference used CANDU fuel (burnup 685 GJ/kg U) as a function of cooling time [5]. Figure 6 shows the relative values of the gamma dose rate as a function of burnup for different cooling periods (1, 10, 100 and 1000 a). These results were obtained using the gamma decay heat data of Smith et al. [20]. One can obtain the value of gamma dose rates for burnup values other than 685 GJ/kg U using the data given in Figures 5 and 6 and following a procedure similar to that described for alpha dose rates (Section 2.1).

The gamma dose rate in water in contact with the used fuel is not only a function of the fuel burnup and cooling period but also of the amount and geometry of the fuel, due to the long "half-thickness value" (the thickness of the absorber medium required to reduce the intensity of the gamma-radiation by one half) for gamma radiation. A water layer in contact with the used fuel receives a gamma dose rate not only from the fuel it is in contact with, but also from the nearby used fuel. Thus, the dose rate near the surface of a used nuclear fuel sample consisting of a few grams will be much smaller than that near the surface of a fuel sample consisting of several kilograms , e.g., a fuel bundle [30].

4. DOSE RATE IN WATER DUE TO NEUTRONS

The neutron dose rate in water in contact with the used CANDU fuel is about two to three orders of magnitude lower than the gamma dose rate for cooling periods up to 10000 a [31]. Therefore, the radiolysis of water by neutrons will not have any significant effect on fuel corrosion in a geological disposal vault. It may be noted here that the G values for water radiolysis due to the neutron dose rate are similar to that for the alpha radiation (i.e., high LET radiation) [32].

5. SUMMARY AND CONCLUSIONS

The report describes procedures to obtain alpha, beta and gamma dose rates in water in contact with the used CANDU fuels of different burnups and cooling times. This information is needed to compare the results of leaching and corrosion experiments [1,5,10,22,30,33] and is also essential to develop models for predicting the oxidation and dissolution rates of the used nuclear fuel in a geological disposal vault, since the radiolysis of water will be the main source of oxidants in the vault [1,10,17].

The results presented here are based on decay heats for alpha, beta and gamma radiations in used CANDU fuel calculated using CANIGEN II computer code [20]. The same procedure can be used to calculate the dose rates from the decay heats obtained using an improved code,

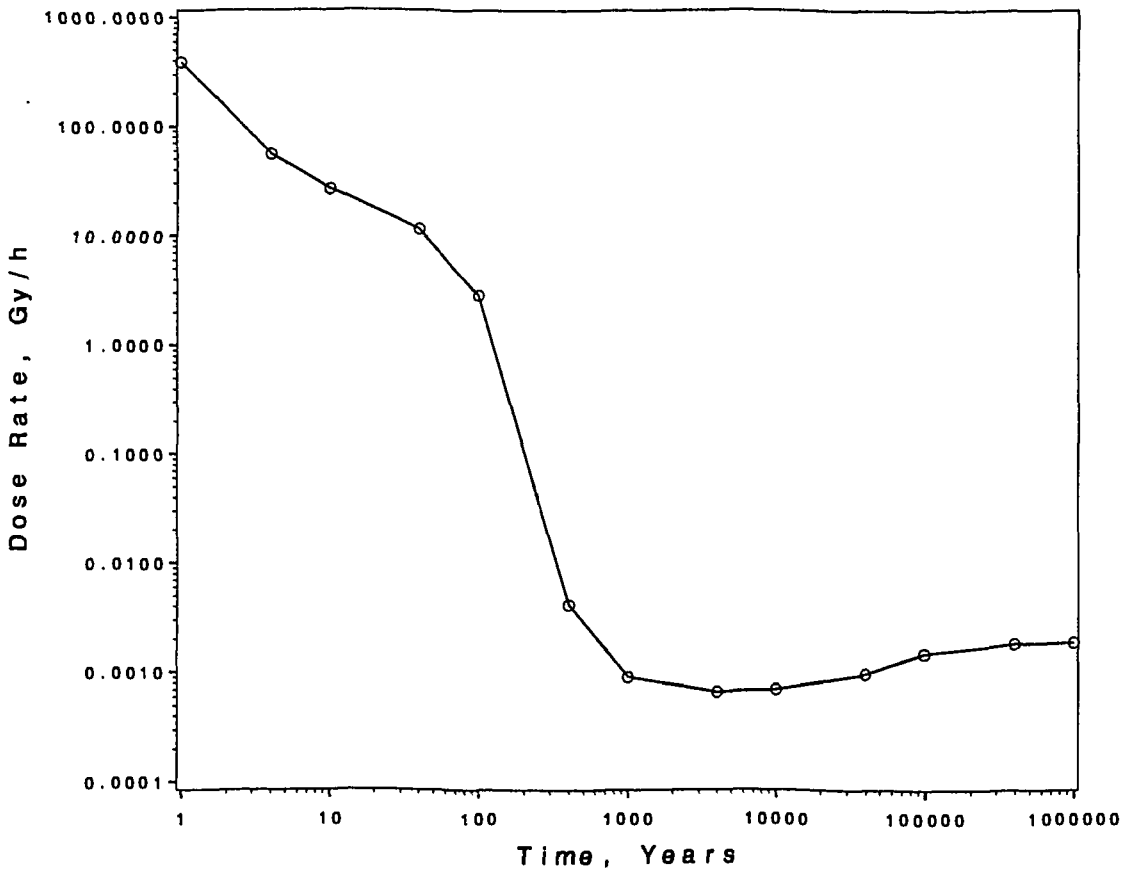

FIGURE 5: The Gamma Dose Rate in Water Near the Surface of a Used CANDU Fuel Bundle, Burnup 685 $GJ \cdot kg^{-1}$ U, as a Function of Cooling Time.

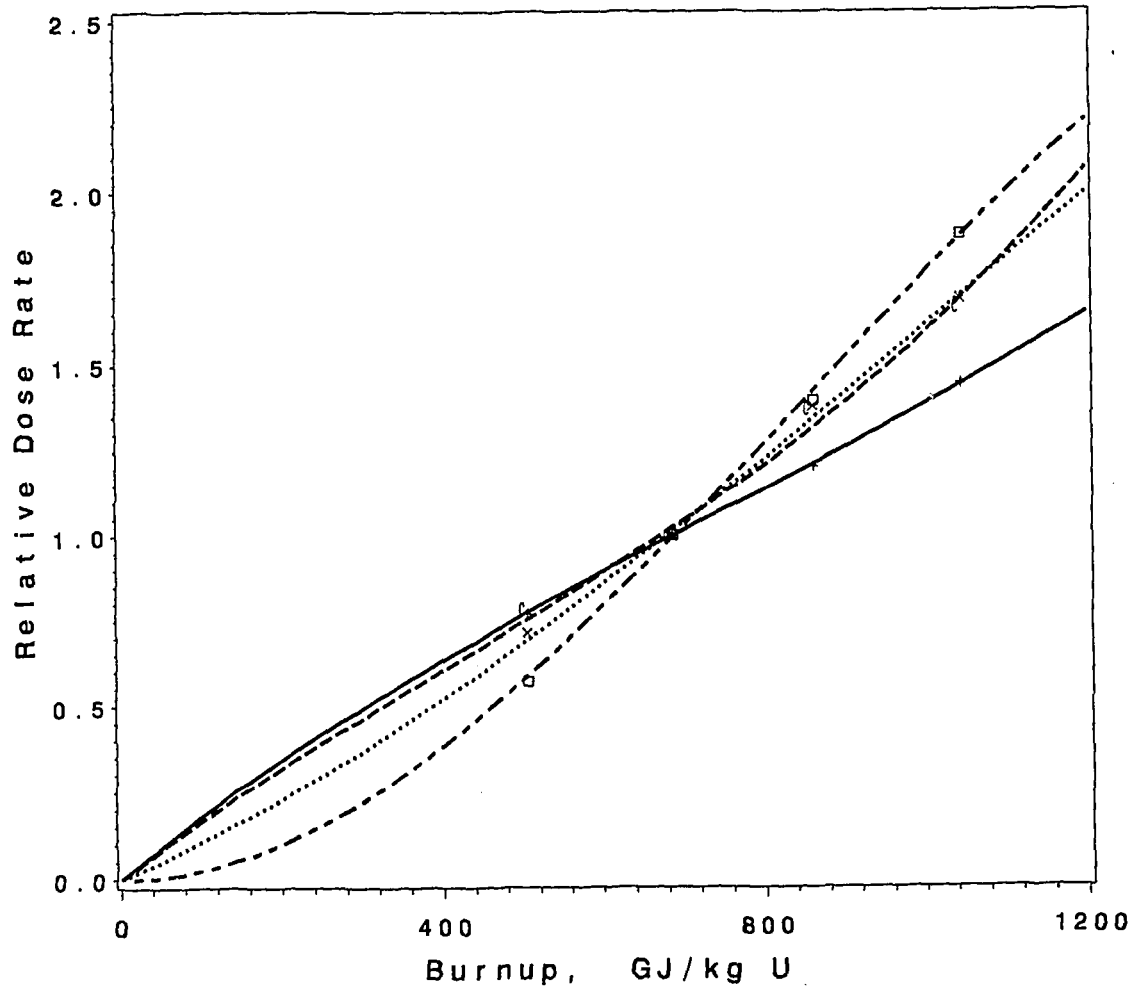

FIGURE 6: The Relative Gamma Dose Rate in Water Near the Surface of Used CANDU Fuel as a Function of Burnup after Different Cooling Periods: (a) 1 a, +; (b) 10 a, X; (c) 100 a, O; and (d) 1000 a, □.

if it were to become available. Also, the procedure can be adapted to estimate dose rates for fuels other than CANDU fuel.

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REFERENCES

1. Johnson, L.H., D.M. Leneveu, D.W. Shoesmith, D.W. Oscarson, M.N. Gray, R.J. Lemire and N.C. Garisto. 1994. The disposal of Canada's nuclear fuel waste: the vault model for postclosure assessment. Atomic Energy of Canada Limited Report, AECL-10714.

2. Grambow, B., A. Loida, P. Dressler, H. Geckeis, P. Diaz, J. Gago, I. Casas, J. Depablo, J. Gimenez and M.E. Torrero. 1994. Long-term safety of radioactive waste disposal: reaction of high burnup spent fuel and UO_2 in saline brines at room temperature. Kernforschungszentrum Karlsruhe Report, KfK 5377.

3. Forsyth, R.S. and L.O. Werme. 1992. Spent fuel corrosion and dissolution. J. Nucl. Mater. 190, 3-19.

4. Gray, W.J., H.R. Leider and S.A. Steward. 1992. Parametric study of LWR spent fuel dissolution kinetics. J. Nucl. Mater. 190, 46-52.

5. Shoesmith, D.W. and S. Sunder. 1992. The prediction of nuclear fuel (UO_2) dissolution rates under waste disposal conditions. J. Nucl. Mater. 190, 20-35.

6. Sunder, S., D.W. Shoesmith, H. Christensen and N.H. Miller. 1992. Oxidation of UO_2 fuel by the products of gamma radiolysis of water. J. Nucl. Mater. 190, 78-86.

7. Matzke, Hj. 1992. Radiation damage-enhanced dissolution of UO_2 in water. J. Nucl. Mater. 190, 101-106.

8. Christensen, H., R.S. Forsyth, R. Lundqwist and L.O. Werme. 1990. Radiation-induced dissolution of UO_2. Studsvik Report, NS-90/85, Studsvik Nuclear, Nykoping, Sweden.

9. Sunder, S., G.D. Boyer and N.H. Miller. 1990. XPS studies of UO_2 oxidation by alpha radiolysis of water at 100°C. J. Nucl. Mater. 175, 163-169.

10. Sunder, S. and D.W. Shoesmith. 1991. Chemistry of UO_2 fuel dissolution in relation to the disposal of used nuclear fuel. Atomic Energy of Canada Limited Report, AECL-10395.

11. Parks, G.A. and D.C. Pohl. 1988. Hydrothermal solubility of uraninite. Geochim. Cosmochim. Acta 52, 863.

12. Lemire, R.J. and F. Garisto. 1989. The solubility of U, Nb, Pu, The and Tc in a geological disposal vault for used nuclear fuel. Atomic Energy of Canada Limited Report, AECL-10009.

13. Allen, A.O. 1961. The radiation chemistry of water and aqueous solutions. D. Van Nostrand Co. Inc., Princeton.

14. Spinks, J.W.T. and R.J. Woods. 1990. An introduction to radiation chemistry, 3rd ed., Wiley-Interscience, New York.

15. Elliot, A.J. 1994. Rate constants and G-values for the simulations of the radiolysis of light water over the range 0-300°C. Atomic Energy of Canada Limited Report, AECL-11073.

16. Christensen, H. and E. Bjergbakke. 1986. Application of CHEMSIMUL for groundwater radiolysis. Nucl. Chem. Waste Manage. 6, 265.

17. Christensen, H. and E. Bjergbakke. 1987. Radiation induced dissolution of uranium dioxide. Materials Research Society Symposium Proceedings 84 (Scientific Basis for Nuclear Waste Management X), 115-122.

18. Sunder, S. and H. Christensen. 1993. Gamma-radiolysis of water solutions relevant to the nuclear fuel waste management program. Nucl. Technology, 104, 403-417.

19. Garisto, F. 1989. The energy spectrum of α-particles emitted from used CANDU™ fuel. Annals of Nuclear Energy, 16, 33-38.

20. Smith, H.J., J.C. Tait and R.E. von Massow. 1987. Radioactive decay properties of Bruce "A" CANDU™ UO_2 fuel and fuel recycle waste. Atomic Energy of Canada Limited Report, AECL-9072.

21. Tait, J.C., I.C. Gauld and G.B. Wilkin. 1989. Derivation of initial radionuclide inventories for the safety assessment of the disposal of used CANDU fuel. Atomic Energy of Canada Limited Report, AECL-9881.

22. Johnson, L.H. and D.W. Shoesmith. 1988. Spent fuel. In Radioactive Waste Forms for the Future (W. Lutze and R.C. Ewing, eds), Elsevier Publishers B.V., 635-698.

23. Ingemansson, T. and J. Elkert. 1991. Model for calculation of absorbed alpha and beta radiation dose to water in contact with highly burnt-up nuclear fuel. ABA Atom Report, Asea Brown Boveri, Sweden, RM-91-23.

24. Nitzki, V. and Hj. Matzke. 1973. Stopping power of 1-9 MeV He^{++} ions in UO_2, (U, Pu)O_2, and ThO_2. Physical Review B.8, 1894-1900.

25. International Commission on Radiation Units and Measurements. 1984. Stopping powers for electrons and positrons. ICRU Report 37.

26. Swallow, A.J. 1960. Radiation chemistry of organic compounds. Pergamon Press Ltd. London.

27. Haissinsky, M. 1964. Nuclear chemistry and its applications. Addison-Wesley Publishing Co. Inc., London.

28. Christensen, H. and S. Sunder. 1993. Evaluation of data from calculations of gamma-radiolysis of aqueous solutions for UO_2 oxidation studies. Studsvik Report, Studsvik/M-93/29, Studsvik Material AB, Nykoping, Sweden.

29. Cross, W.G., N.O. Freedman and P.Y. Wong. 1992. Tables of beta-ray dose distribution in water. Atomic Energy of Canada Limited Report, AECL-10521.

30. Shoesmith, D.W., S. Sunder, M.G. Bailey and N.H. Miller. 1995. Corrosion of used nuclear fuel in aqueous perchlorate and carbonate solutions. J. Nucl. Mater. (submitted).

31. Wilkin, G.B. (unpublished results).

32. Bjergbakke, K. Sehested, O.L. Rasmussen and H. Christensen. 1984. Input files for computer simulation of water radiolysis. Riso National Laboratory, RISO-M-2430.

33. Stroes-Gascoyne, S., J.C. Tait, N.C. Garisto, R.J. Porth, J.P.M. Ross, G.A. Glowa and T.R. Barnsdale. 1992. Instant release of ^{14}C, ^{99}C, ^{90}Sr and ^{137}Cs from used CANDU fuel at 25°C in distilled deionized water. Materials Research Society Symposium Proceedings 257 (Scientific Basis for Nuclear Waste Management XV), 373-380.

APPENDIX A

CALCULATION OF ALPHA DOSE RATE IN WATER IN CONTACT WITH USED CANDU FUEL FROM GARISTO'S RESULTS

Garisto has calculated the energy emitted, as alpha particles, by a unit area of used fuel (E_{out}, $MeV \cdot cm^{-2} \cdot s^{-1}$) for four different CANDU fuels [A1]. One can calculate the dose rate in water from E_{out} assuming that the emitted alpha-particle energy is absorbed in the water layer of thickness, d, equal to the range of alpha particles in water.

Garisto has also described a procedure to calculate the range of alpha particles in water. Figure A1 shows the range of alpha particles in water as a function of their energy. It compares the range calculated using Garisto's procedure with the literature values [A2,A3]. Garisto suggests that one can calculate the alpha-particle range in water using the equation

$$R = \frac{A_w}{2} \cdot E_o^2 + B_w \cdot E_o \qquad \text{(A.1)}$$

where R = range of alpha particle, μm; E_o = alpha-particle energy in the fuel, MeV; and A_w and B_w are constants derived using the data of Whaling (A4). During the calculation of the range of alpha particles (using Garisto's procedure) it became apparent that the values of A_w and B_w in Garisto's paper were interchanged. The correct values are $A_w = 1.60$ $\mu m \cdot MeV^{-2}$ and $B_w = 2.45$ $\mu m \cdot MeV^{-1}$. (This point has been discussed with F. Garisto.) The alpha dose rates in water obtained using the E_{out} values given in Table A1 are shown in Table 2 of the main text for two different values of water layer thickness.

TABLE A1

GARISTO'S RESULTS ON THE ALPHA PARTICLES EMITTED FROM FOUR USED CANDU FUELS

Used Fuel #	Burnup	Cooling Time	E_{out}*
	$GJ \cdot kg^{-1}$ U	a	$MeV \cdot cm^{-2} \cdot s^{-1}$
1	685	100	3.44×10^5
2	685	1000	1.63×10^5
3	685	10000	6.16×10^4
4	1045	1000	2.43×10^5

* Alpha particle energy emitted per unit area of the fuel.

REFERENCES

A1. Garisto, F. 1989. The energy spectrum of α-particles emitted from used CANDU fuel. Annals of Nuclear Energy 16, 33-38.

A2. Haissinsky, M. 1964. Nuclear Chemistry and Its Applications. Addison-Wesley Publishing Co. Inc. London.

A3. Spinks, J.W.T. and R.J. Woods. 1990. An Introduction to Radiation Chemistry, 3rd Ed. Wiley-Interscience, New York.

A4. Waling, W. 1958. The Energy Loss of Charged Particles in Matter. In Encyclopedia of Physics, Vol.XXXIV), (S. Flugge, editor), Springer Verlag, Berlin, pp.193-217.

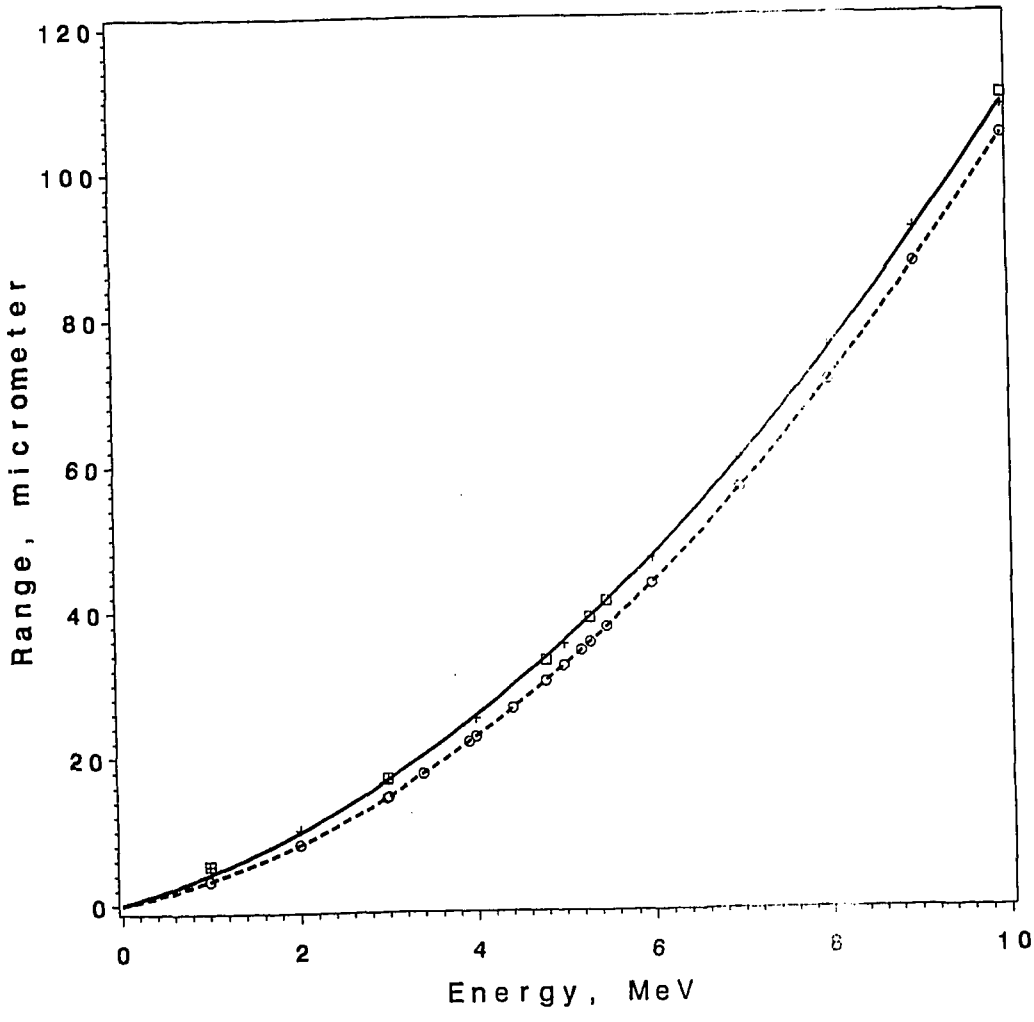

FIGURE A-1: Range of Alpha-Particles in Water as a Function of Energy: (a) using Garisto's equation with correct parameters (see text), ○ (dashed line); (b) Haissinsky's data, + (solid line); and (c) Spinks and Wood's data, □.

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