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Summary

In cooperation with ITU, Germany, SKB has used alpha-doped fuel pellets produced by mixing alpha-active U-233 into a depleted UO_2 pellet. The aim has been to produce a fuel pellet that is representative, from a radiation point of view, of a reactor fuel pellet with typical final burnup and a decay period of typically 3 000 years.

The purpose of the present study was to verify that the levels of alpha, electron and gamma radiation for the U-233 pellet are similar to the decayed reactor fuel pellet. A calculation of the radiation source terms, expressed in W/kg fuel pellet, for the U-233 pellet and the reactor fuel pellet was therefore made. The reactor fuel pellet has been selected from a BWR fuel assembly with a burnup of 60 MWd/kgU. Decay periods of 1 000, 3 000 and 10 000 years have been selected for the reactor fuel. Based on the calculated radiation source terms, dose rates, expressed in Gy/h for alpha, electron and gamma radiation, have been estimated for the water at the water/pellet interface, and average radiation doses in a water layer of typically 34 µm, which is the estimated range for the alpha radiation.

A summary of estimated dose rates to water outside the U-233 doped pellet and the reactor pellet with a burnup of 60 MWd/kgU (BU60 pellet) is presented in the below table. Some comments and conclusions:

- The average alpha dose rate in a water layer up to 34 µm is about a factor 8 lower than the alpha dose rate at the interface pellet-water.
- The average electron and gamma dose rates are estimated to be quite constant within the alpha 34 µm range.
- The average alpha dose rates are significantly higher than the electron dose rates (a factor of 110 to 270) and the gamma dose rates (a factor of 2 000 to 5 600) both for the alpha-doped pellet and spent fuel.
- All dose rates for the U-233 doped pellet are quite similar to the dose rates for the BU60 pellet at the considered decay periods of 1 000, 3 000 and 10 000 years.
  - The electron dose rate for the alpha-doped pellet is ~ 0.4 % of the average alpha dose rate, while in the case of the 3 000 years old fuel it is ~ 0.9 % of the average alpha dose rate. The gamma dose rate is ~ 0.033 % of the average alpha dose rate for the alpha-doped pellet and ~ 0.030 % for the 3 000 years old fuel.

U-233 and BU60 pellet (1 000, 3 000 and 10 000 years) – Dose rate to water in contact – Summary.

	Alpha dose rate in water, Gy/h		Electron dose rate in water, Gy/h		Gamma dose rate in water, Gy/h	
	Max at UO_2 surface	Average 0–34 µm	Max at UO_2 surface	Average 0–34 µm	Max at UO_2 surface	Average 0–34 µm
U-233 doped pellet	680	82.8	0.31	0.31	0.027	0.027
BU60 pellet, decay 1 000 y	1 111	135	1.23	1.23	0.024	0.024
BU60 pellet, decay 3 000 y	499	61	0.54	0.54	0.018	0.018
BU60 pellet, decay 10 000 y	281	34	0.31	0.31	0.017	0.017

Sammanfattning

I samarbete med ITU, Tyskland har SKB använt alfa-dopade bränslekutsar framställda genom att blanda alfa-aktiv U-233 i en utarmad UO_2-kuts. Syftet har varit att producera en bränslekuts som är representativ, från strålningssynpunkt, för en reaktorbränslekuts med typisk slututbränning och en avklingningstid på vanligen 3000 år.

Syftet med föreliggande studie var att verifiera att nivåerna av alfa-, elektron- och gammastrålning för U-233-pelleten liknar den utbrända reaktorbränslekutsen. En beräkning av källtermer, uttryckt i W/kg bränslekuts, för U-233-kutsen och reaktorbränslekutsen gjordes därför. Reaktorbränslekutsen har valts från en BWR-bränslepatron med utbränning 60 MWd / kgU. Avklingningstider på 1 000, 3 000 och 10 000 år har valts ut för reaktorbränslet. Baserat på källtermsberäkningarna har dos till vatten, uttryckt i Gy/h för alfa-, elektron- och gammastrålning, uppskattats precis vid ytan av kutsen samt i ett omgivande vattenskikt på typiskt 34 µm, vilket motsvarar alfa-strålningens räckvidd.

En sammanfattning av beräknade dosrater för vatten utanför den U-233-dopade kutsen och reaktorbränslekutsen presenteras i nedanstående tabell. Några kommentarer och slutsatser:

- Den genomsnittliga alfadosen i vattenskiktet upp till 34 µm är ungefär en faktor 8 lägre än alfadosen vid gränssnittet mellan kuts och vatten.
- De genomsnittliga elektron- och gamma-doserna uppskattas vara ganska konstanta inom alfastrålningens räckvidd, 34 µm.
- De genomsnittliga alfadoserna är signifikant högre än elektrondoserna (med en faktor 110 till 270) och gammadoserna (med en faktor 2 000 till 5 600), både för alfa-dopad kuts och använt bränsle.
- Samtliga dosrater för U-233-dopad kuts är jämförbara med dosraterna för BU60-kutsen vid de bedömda avklingningstiderna 1 000, 3 000 och 10 000 år.
  - Elektrondosraten för den alfa-dopade kutsen är ~ 0,4 % av den genomsnittliga alfadosraten, medan den för ett 3000 år gammalt bränsle är ~ 0,9 % av den genomsnittliga alfadosraten. Gammadosen är ~ 0,033 % av den genomsnittliga alfadosraten för den alfa-dopade kutsen och ~ 0,030 % för det 3 000-åriga bränslet.

U-233 och BU60 kutsen (1 000, 3 000 och 10 000 år) – dosrat för vatten i kontakt med kuts – sammanfattning.

	Alfadosrat i vatten, Gy/h		Elektrondosrat i vatten, Gy/h		Gammadosrat i vatten, Gy/h	
	Max vid UO_2-ytan	Medel 0–34 µm	Max vid UO_2-ytan	Medel 0–34 µm	Max vid UO_2-ytan	Medel 0–34 µm
U-233 dopad kuts	680	82.8	0.31	0.31	0.027	0.027
BU-60 kuts, avklingning 1 000 år	1 111	135	1.23	1.23	0.024	0.024
BU-60 kuts, avklingning 3 000 år	499	61	0.54	0.54	0.018	0.018
BU-60 kuts, avklingning 10 000 år	281	34	0.31	0.31	0.017	0.017

Contents

1 Introduction

In cooperation with ITU, Germany, SKB has used alpha-doped fuel pellets produced by mixing alpha-active U-233 into a depleted UO_2 pellet. The aim has been to produce a fuel pellet that is representative, from a radiation point of view, of a reactor fuel pellet with typical final burnup and a decay period of typically 3 000 years.

The purpose of the present study was to verify that the levels of alpha, electron[1] and gamma radiation for the U-233 pellet are similar to the decayed reactor fuel pellet. A calculation of the radiation source terms, expressed in W/kg fuel pellet, for the U-233 pellet and the reactor fuel pellet was therefore made. The reactor fuel pellet has been selected from a BWR fuel assembly with a burnup of 60 MWd/kgU. Decay periods of 1 000, 3 000 and 10 000 years have been selected for the reactor fuel. Based on the calculated radiation source terms, dose rates, expressed in Gy/h for alpha, electron and gamma radiation, have been estimated for the water at the water/pellet interface, and average radiation doses in a water layer of typically 34 µm, which is the estimated range for the alpha radiation.

The result of the calculations is presented in the report.

[1] With "electron radiation" in this report is meant all type of radiation involving electrons, i.e. beta radiation, as well as Auger and conversion electrons. In Figure 2-1 and Figure 2-2, the symbol "beta" represents "electron radiation".

2 Radiation source terms

2.1 U-233 doped pellet

The radioactivity in the U-233 doped pellet has been measured (Carbol et al. 2005, Table 22). The measured activity is presented in Table 2-1. The activity stems from several decay schemes:

- The main activity is due to U-233 with daughters, see Figure 2-1.
- Some is due to U-232 with daughters, see Figure 2-2.
- Some is due to other U isotopes (U-234, U-235, U-236 and U-238).
- Some is due to trace amounts of actinide nuclides (Pu-238, Pu-239, Pu-240, Pu-241, Pu-242 and Am-241).

Table 2-1. Measured radioactivity in 10 % U-233 doped UO_2 pellet (Carbol et al. 2005, Table 22).

Radionuclide	Decay mode	$t_{1/2}$	A, 10 % (Bq/g UO_2-pellet)
^{232}U-chain			
^{232}U	α	69.85 y	7.898×10^4
^{228}Th	α	1.914 y	8.120×10^4
^{224}Ra	α	3.62 d	8.122×10^4
^{220}Rn	α	55.6 s	8.122×10^4
^{216}Po	α	145 ms	8.122×10^4
^{212}Pb	β	10.64 h	8.122×10^4
^{212}Bi	β(α)	1.009 h	8.122×10^4
^{212}Po	α	300 ns	8.122×10^4
^{208}Tl	β	3.055 min	29190
^{233}U-chain			
^{233}U	α	1.594×10^5 y	3.605×10^7
^{229}Th	α	7.345×10^3 y	1.291×10^5
^{225}Ra	β	14.80 d	1.289×10^5
^{225}Ac	α	10.0 d	1.287×10^5
^{221}Fr	α	4.9 min	1.287×10^5
^{217}At	α(β)	32.3 ms	1.287×10^5
^{217}Rn	α	540 µs	1.287×10^5
^{213}Po	α	4.2 µs	1.287×10^5
^{213}Bi	β(α)	5.59 min	1.287×10^5
^{209}Pb	β	3.253 min	1.287×10^5
^{209}Tl	β	2.2 min	2780
^{234}U	α	2.484×10^5 y	6.22×10^4
^{235}U	α	7.038×10^8 y	263
^{236}U	α	2.34×10^7 y	411
^{238}U	α	4.699×10^9 y	1.06×10^4
^{238}Pu	α	87.744 y	5.2×10^5
^{239}Pu	α	2.413×10^4 y	4.15×10^5
^{240}Pu	α	6.537×10^3 y	2.32×10^5
^{239,240}Pu	α		4.6×10^5
^{241}Pu	β	15.16 y	6.9×10^6
^{242}Pu	α	3.869×10^5 y	436
^{241}Am	α	433.176 y	4.3×10^5

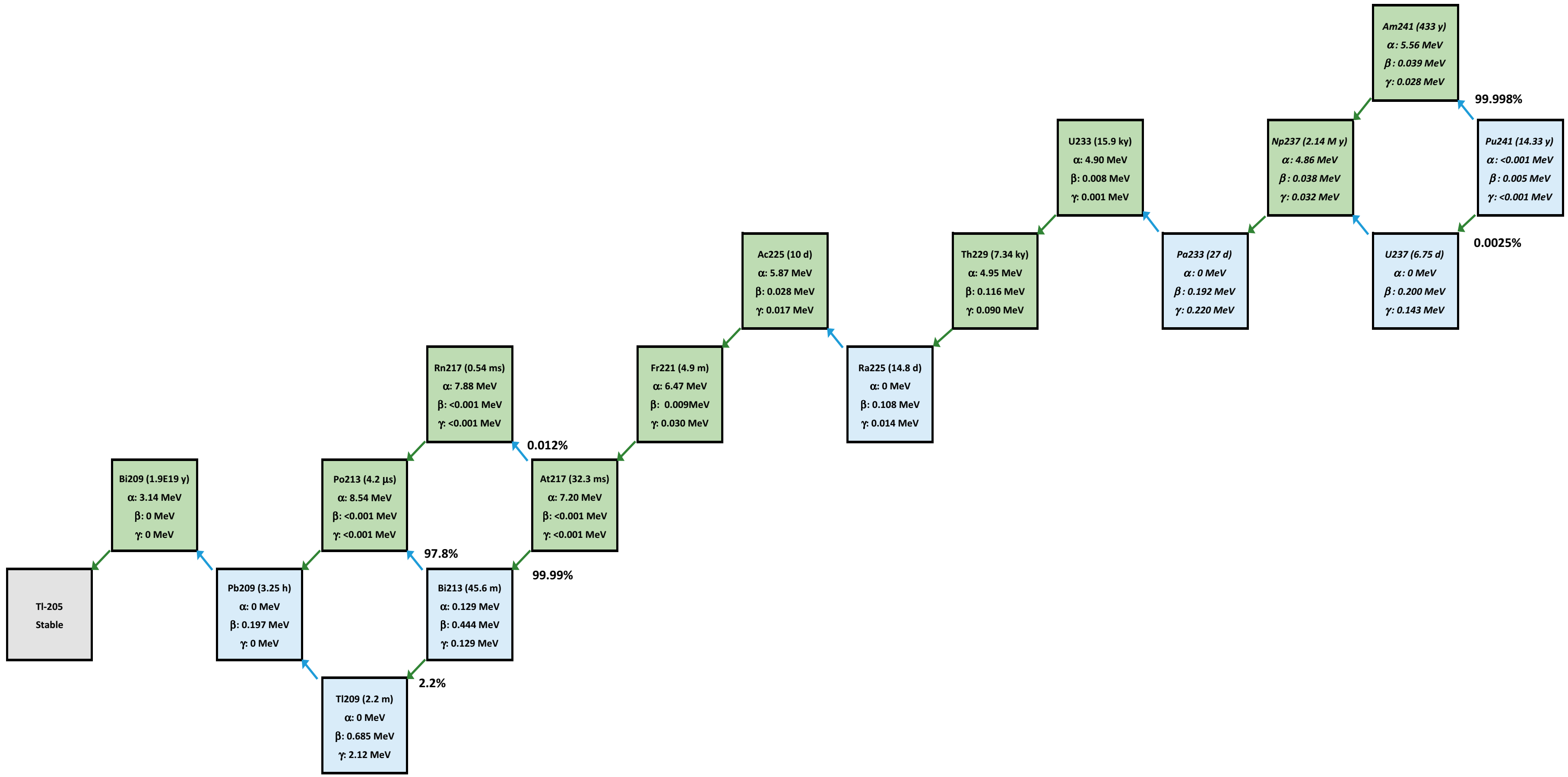

Figure 2-1. *The U-233 decay chain with radiation yields (Forrest 2007).*

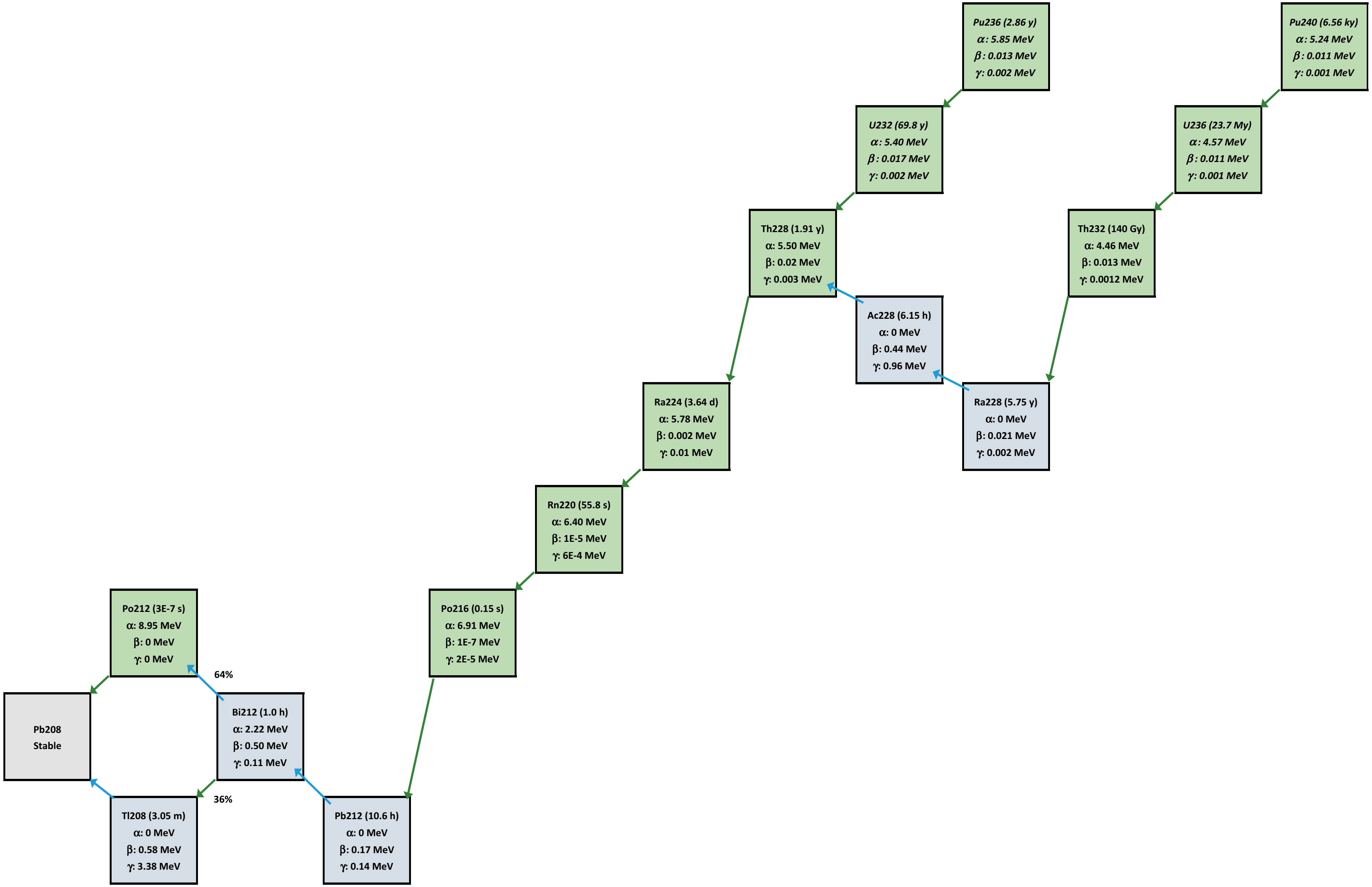

Figure 2-2. *The U-232 decay chain with radiation yields (Forrest 2007).*

Based on the radioactivity presented in Table 2-1 the radiation source terms in the U-233 pellet are calculated using the FISPACT-II code (Sublet et al. 2015). In order to determine the dependence of the source term strengths on the decay period, strengths for six different periods between 0.01 and 1 000 years have been calculated. The results for alpha, electron and gamma source terms are presented in Table 2-2. The radiation source term is very dominated by the alpha contribution and is rather unaffected by decay period up to at least 100 years for the total and alpha doses and up to at least 10 years for electron and gamma doses.

Table 2-2. U-233 doped pellet – Radiation source terms expressed as W/kg fuel pellet vs. decay period.

Decay [y]	Source term [W/kg]		
	Alpha	Electron	Gamma
0.01	3.09E–02	8.54E–05	3.53E–05
0.1	3.09E–02	8.62E–05	3.53E–05
1	3.09E–02	8.69E–05	3.53E–05
10	3.11E–02	8.88E–05	3.56E–05
100	3.22E–02	1.23E–04	4.13E–05
1 000	4.66E–02	5.40E–04	1.83E–04

The radiation source term contributions from different nuclides are presented in Table 2-3. The follow nuclides are dominating:

- Alpha[2]: U-233, ca 91 %. The U-233 alpha energy is 4.9 MeV/dis., see Figure 2-1.
- Gamma: Tl-208, ca 45 %, and U-233, ca 20 %. The average gamma energy for Tl-208 is 3.38 MeV/dis. (Figure 2-2), to be compared to only 0.001 MeV/dis. for U-233 (Figure 2-1).
- Electron: U-233, ca 51 %, Bi-213, ca 11 %, Bi-212, ca 8 % and Pu-241, ca 7 %. The average electron energies are 0.008, 0.444, 0.503 and 0.005 MeV/dis., respectively (Forrest 2007).

Table 2-3. U-233 doped pellet – Decay 0.01 years – Radiation source terms expressed as W/kg fuel pellet – Dominant nuclides.

Nuclide	Total heat (W/kg)	Percent Heat	Nuclide	Gamma heat (W/kg)	Percent Gamma	Nuclide	Electron heat (W/kg)	Percent Electron
Total	3.10E–02		Total	3.53E–05		Total	8.54E–05	
U-233	2.83E–02	9.13E+01	Tl208	1.58E–05	4.49E+01	U-233	4.38E–05	5.13E+01
Pu238	4.67E–04	1.50E+00	U-233	7.07E–06	2.00E+01	Bi213	9.15E–06	1.07E+01
Am241	3.87E–04	1.25E+00	Bi213	2.66E–06	7.54E+00	Bi212	6.54E–06	7.67E+00
Pu239	3.49E–04	1.13E+00	Am241	1.90E–06	5.37E+00	Pu241	5.78E–06	6.77E+00
Pu240	1.95E–04	6.30E–01	Pb212	1.88E–06	5.34E+00	Pb209	4.06E–06	4.76E+00
Po213	1.72E–04	5.54E–01	Th229	1.87E–06	5.29E+00	Tl208	2.74E–06	3.20E+00
At217	1.48E–04	4.78E–01	Bi212	1.40E–06	3.96E+00	Am241	2.69E–06	3.15E+00
Fr221	1.34E–04	4.32E–01	Tl209	9.44E–07	2.68E+00	Th229	2.40E–06	2.81E+00
Ac225	1.22E–04	3.92E–01	Fr221	6.14E–07	1.74E+00	Ra225	2.23E–06	2.61E+00
Th229	1.07E–04	3.43E–01	Ac225	3.53E–07	1.00E+00	Pb212	2.20E–06	2.57E+00
Po216	8.99E–05	2.90E–01	Ra225	2.86E–07	8.10E–01	Pu238	9.34E–07	1.09E+00
Rn220	8.34E–05	2.69E–01	Ra224	1.36E–07	3.85E–01	Ac225	5.69E–07	6.66E–01
Ra224	7.54E–05	2.43E–01	Pu238	1.29E–07	3.66E–01	Pu239	5.28E–07	6.19E–01
Po212	7.47E–05	2.41E–01	Pu240	5.06E–08	1.44E–01	Pu240	4.13E–07	4.84E–01
Th228	7.18E–05	2.32E–01	Pu239	4.84E–08	1.37E–01	Tl209	3.05E–07	3.57E–01
U-232	6.85E–05	2.21E–01	Th228	4.00E–08	1.14E–01	Th228	2.66E–07	3.12E–01
U-234	4.84E–05	1.56E–01	U-232	2.13E–08	6.05E–02	U-232	2.13E–07	2.50E–01
Bi212	3.68E–05	1.19E–01	U-234	1.45E–08	4.10E–02	Fr221	1.82E–07	2.13E–01
Tl208	1.86E–05	5.98E–02	Rn220	8.18E–09	2.32E–02	U-234	1.41E–07	1.65E–01
Bi213	1.45E–05	4.66E–02	U-235	6.90E–09	1.96E–02	Pa234m	1.38E–07	1.62E–01
Rest	2.61E–05	8.42E–02	Rest	1.81E–08	5.15E–02	Rest	7.69E–08	9.01E–02

[2] Approximately equal "Heat" in Table 2-3 which actually also includes electron and gamma contributions.

2.2 Irradiated pellet

The radioactivity for a reactor fuel pellet after burnup is taken from a calculation used for shielding calculations for the copper canisters in Clink (Lundgren 2012):

- BWR fuel assembly with average burnup 60 MWd/kgU. Central fuel pellet position with somewhat higher burnup level, 67 MWd/kgU, has been selected.
- The radioactivity has been recalculated to alpha, electron and gamma radiation source terms, for the three decay periods 1 000, 3 000 and 10 000 years with the computer code FISPACT-II (Sublet et al. 2015). The calculation results are presented in Table 2-4. The source terms are quite similar to those for the U-233 pellet, see Table 2-2. Some influence of the decay period can be seen.

Table 2-4. BWR 60 MWd/kgU pellet – radiation source terms expressed as W/kg fuel pellet vs. decay period.

	Source term [W/kg]		
Decay [y]	Alpha	Electron	Gamma
1 000	5.05E–02	3.42E–04	2.25E–04
3 000	2.27E–02	1.50E–04	9.22E–05
10 000	1.28E–02	8.68E–05	5.33E–05

The dominant nuclides for the decay periods 1 000, 3 000 and 10 000 years are shown in Table 2-5, Table 2-6 and Table 2-7, respectively. The following nuclides are dominating for the decay period 3 000 years:

- Alpha[3]: Pu-240 (61 %) and Pu-239 (25 %).
- Gamma: Np-239 (57 %) and Am-243 (18 %).
- Electron: Np 239 (51 %) and Pu-240 (20 %).

Table 2-5. BWR 60 MWd/kgU pellet – Decay 1 000 years – Activity and radiation source terms expressed as W/kg fuel pellet – Dominant nuclides.

Nuclide	Activity (Bq/kg)	Percent Activity	Nuclide	Total Heat (W/kg)	Percent Heat	Nuclide	Gamma heat (W/kg)	Percent Gamma	Nuclide	Electron Heat (W/kg)	Percent Electron
Total	6.91E+10		Total	5.11E–02		Total	2.25E–04		Total	3.42E–04	
Am241	2.80E+10	4.05E+01	Am241	2.52E–02	4.94E+01	Am241	1.23E–04	5.48E+01	Am241	1.75E–04	5.12E+01
Pu240	2.06E+10	2.98E+01	Pu240	1.73E–02	3.39E+01	Np239	6.32E–05	2.81E+01	Np239	9.12E–05	2.66E+01
Pu239	7.19E+09	1.04E+01	Pu239	6.05E–03	1.18E+01	Am243	1.98E–05	8.77E+00	Pu240	3.66E–05	1.07E+01
U-235m	7.19E+09	1.04E+01	Am243	1.89E–03	3.70E+00	Sb126m	8.75E–06	3.88E+00	Tc 99	1.14E–05	3.32E+00
Np239	2.17E+09	3.13E+00	Np239	1.54E–04	3.02E–01	Pu240	4.49E–06	1.99E+00	Pu239	9.14E–06	2.67E+00
Am243	2.17E+09	3.13E+00	Pu242	1.43E–04	2.81E–01	Sb126	2.16E–06	9.58E–01	Am243	8.30E–06	2.43E+00
Tc 99	8.30E+08	1.20E+00	U-234	8.67E–05	1.70E–01	Pa233	1.38E–06	6.13E–01	Sb126m	3.47E–06	1.01E+00
Pu242	1.80E+08	2.60E–01	Pu238	8.66E–05	1.70E–01	Pu239	8.39E–07	3.72E–01	Pa234m	1.31E–06	3.81E–01
Zr 93	1.28E+08	1.85E–01	Np237	3.09E–05	6.05E–02	Cm245	4.05E–07	1.80E–01	Pa233	1.20E–06	3.52E–01
Nb 93m	1.24E+08	1.80E–01	Cm245	2.43E–05	4.75E–02	Sn126	3.13E–07	1.39E–01	Sn126	6.07E–07	1.77E–01
U-234	1.11E+08	1.61E–01	Cm246	1.28E–05	2.51E–02	Np237	2.03E–07	9.01E–02	Nb 93m	5.74E–07	1.68E–01
Pu238	9.67E+07	1.40E–01	Sb126m	1.22E–05	2.39E–02	Ho166m	1.11E–07	4.94E–02	Zr 93	3.93E–07	1.15E–01
Np237	3.91E+07	5.65E–02	U-236	1.17E–05	2.29E–02	Nb 93m	4.08E–08	1.81E–02	Cs135	3.56E–07	1.04E–01
Pa233	3.91E+07	5.65E–02	Tc 99	1.14E–05	2.22E–02	Bi214	4.00E–08	1.78E–02	Cm245	3.51E–07	1.03E–01
Sb126m	3.47E+07	5.01E–02	U-238	6.83E–06	1.34E–02	Pu242	3.72E–08	1.65E–02	C 14	2.74E–07	7.99E–02
Sn126	3.47E+07	5.01E–02	Cm242	2.94E–06	5.76E–03	Pa234m	3.16E–08	1.40E–02	Pu242	2.70E–07	7.89E–02
C 14	3.45E+07	4.99E–02	Pa233	2.58E–06	5.06E–03	U-234	2.59E–08	1.15E–02	Sb126	2.62E–07	7.65E–02
Cs135	2.93E+07	4.24E–02	Sb126	2.42E–06	4.74E–03	Pu238	2.40E–08	1.06E–02	U-234	2.52E–07	7.38E–02
Pu241	2.70E+07	3.90E–02	Pa234m	1.34E–06	2.62E–03	Th234	1.41E–08	6.25E–03	Np237	2.40E–07	7.01E–02
Cm245	2.69E+07	3.90E–02	Sn126	9.20E–07	1.80E–03	Am242	9.46E–09	4.20E–03	Pu238	1.73E–07	5.06E–02
Rest	1.21E+08	1.75E–01	Rest	4.16E–06	8.15E–03	Rest	4.28E–08	1.90E–02	Rest	6.28E–07	1.83E–01

[3] Approximately equal "Heat" in Table 2-5, Table 2-6 and Table 2-7 which actually also includes electron and gamma contributions.

Table 2-6. BWR 60 MWd/kgU pellet – Decay 3 000 years – Activity and radiation source terms expressed as W/kg fuel pellet – Dominant nuclides.

Nuclide	Activity (Bq/kg)	Percent Activity	Nuclide	Total Heat (W/kg)	Percent Heat	Nuclide	Gamma Heat (W/kg)	Percent Gamma	Nuclide	Electron Heat (W/kg)	Percent Electron
Total	3.69E+10		Total	2.29E–02		Total	9.22E–05		Total	1.50E–04	
Pu240	1.67E+10	4.51E+01	Pu240	1.40E–02	6.12E+01	Np239	5.24E–05	5.68E+01	Np239	7.55E–05	5.05E+01
Pu239	6.90E+09	1.87E+01	Pu239	5.80E–03	2.53E+01	Am243	1.64E–05	1.78E+01	Pu240	2.97E–05	1.98E+01
U-235m	6.90E+09	1.87E+01	Am243	1.56E–03	6.82E+00	Sb126m	8.70E–06	9.44E+00	Tc 99	1.13E–05	7.53E+00
Np239	1.79E+09	4.86E+00	Am241	1.05E–03	4.57E+00	Am241	5.13E–06	5.56E+00	Pu239	8.77E–06	5.86E+00
Am243	1.79E+09	4.86E+00	Pu242	1.43E–04	6.24E–01	Pu240	3.64E–06	3.94E+00	Am241	7.28E–06	4.86E+00
Am241	1.16E+09	3.15E+00	Np239	1.28E–04	5.58E–01	Sb126	2.15E–06	2.33E+00	Am243	6.88E–06	4.59E+00
Tc 99	8.24E+08	2.23E+00	U-234	8.63E–05	3.77E–01	Pa233	1.57E–06	1.71E+00	Sb126m	3.45E–06	2.30E+00
Pu242	1.79E+08	4.85E–01	Np237	3.52E–05	1.53E–01	Pu239	8.05E–07	8.73E–01	Pa233	1.37E–06	9.15E–01
Zr 93	1.28E+08	3.46E–01	Cm245	2.06E–05	9.00E–02	Cm245	3.44E–07	3.73E–01	Pa234m	1.31E–06	8.72E–01
Nb 93m	1.24E+08	3.36E–01	U-236	1.25E–05	5.45E–02	Bi214	3.13E–07	3.40E–01	Sn126	6.03E–07	4.03E–01
U-234	1.11E+08	3.00E–01	Sb126m	1.21E–05	5.30E–02	Sn126	3.11E–07	3.38E–01	Nb 93m	5.73E–07	3.83E–01
Np237	4.45E+07	1.21E–01	Tc 99	1.13E–05	4.92E–02	Np237	2.31E–07	2.51E–01	Zr 93	3.93E–07	2.62E–01
Pa233	4.45E+07	1.21E–01	Cm246	9.58E–06	4.18E–02	Pb214	4.83E–08	5.24E–02	Cs135	3.56E–07	2.38E–01
Sb126m	3.45E+07	9.33E–02	U-238	6.83E–06	2.98E–02	Nb 93m	4.08E–08	4.42E–02	Cm245	2.98E–07	1.99E–01
Sn126	3.45E+07	9.33E–02	Pa233	2.94E–06	1.28E–02	Pu242	3.70E–08	4.02E–02	Np237	2.73E–07	1.82E–01
Cs135	2.93E+07	7.93E–02	Sb126	2.41E–06	1.05E–02	Ho166m	3.50E–08	3.80E–02	Pu242	2.69E–07	1.80E–01
C 14	2.71E+07	7.32E–02	Th230	2.24E–06	9.76E–03	Pa234m	3.16E–08	3.42E–02	Sb126	2.60E–07	1.74E–01
Sb126n	2.30E+07	6.24E–02	Po214	1.59E–06	6.96E–03	U-234	2.58E–08	2.79E–02	U-234	2.51E–07	1.68E–01
Pu241	2.29E+07	6.21E–02	Pa234m	1.34E–06	5.84E–03	Th234	1.41E–08	1.53E–02	C 14	2.14E–07	1.43E–01
Cm245	2.29E+07	6.19E–02	Po218	1.24E–06	5.43E–03	I 129	6.64E–09	7.21E–03	Bi214	1.28E–07	8.54E–02
Rest	9.25E+07	2.51E–01	Rest	7.70E–06	3.36E–02	Rest	3.01E–08	3.26E–02	Rest	5.85E–07	3.91E–01

Table 2-7. BWR 60 MWd/kgU pellet – Decay 10 000 years – Activity and radiation source terms expressed as W/kg fuel pellet – Dominant nuclides.

Nuclide	Activity (Bq/kg)	Percent Activity	Nuclide	Total Heat (W/kg)	Percent Heat	Nuclide	Gamma Heat (W/kg)	Percent Gamma	Nuclide	Electron Heat (W/kg)	Percent Electron
Total	2.33E+10		Total	1.29E–02		Total	5.33E–05		Total	8.68E–05	
Pu240	7.95E+09	3.41E+01	Pu240	6.69E–03	5.19E+01	Np239	2.71E–05	5.08E+01	Np239	3.91E–05	4.50E+01
Pu239	5.88E+09	2.52E+01	Pu239	4.94E–03	3.83E+01	Sb126m	8.52E–06	1.60E+01	Pu240	1.42E–05	1.63E+01
U-235m	5.88E+09	2.52E+01	Am243	8.09E–04	6.27E+00	Am243	8.47E–06	1.59E+01	Tc 99	1.10E–05	1.27E+01
Np239	9.28E+08	3.98E+00	Pu242	1.41E–04	1.09E+00	Sb126	2.10E–06	3.94E+00	Pu239	7.47E–06	8.61E+00
Am243	9.28E+08	3.98E+00	U-234	8.48E–05	6.57E–01	Bi214	1.84E–06	3.45E+00	Am243	3.56E–06	4.10E+00
Tc 99	8.06E+08	3.46E+00	Np239	6.62E–05	5.13E–01	Pu240	1.74E–06	3.26E+00	Sb126m	3.38E–06	3.89E+00
Pu242	1.77E+08	7.58E–01	Np237	3.53E–05	2.74E–01	Pa233	1.58E–06	2.96E+00	Pa233	1.38E–06	1.59E+00
Zr 93	1.27E+08	5.47E–01	U-236	1.43E–05	1.10E–01	Pu239	6.85E–07	1.29E+00	Pa234m	1.31E–06	1.50E+00
Nb 93m	1.24E+08	5.30E–01	Am241	1.24E–05	9.61E–02	Sn126	3.05E–07	5.72E–01	Bi214	7.52E–07	8.66E–01
U-234	1.09E+08	4.67E–01	Sb126m	1.19E–05	9.22E–02	Pb214	2.84E–07	5.32E–01	Sn126	5.91E–07	6.81E–01
Pa233	4.47E+07	1.92E–01	Cm245	1.16E–05	9.03E–02	Np237	2.32E–07	4.35E–01	Nb 93m	5.72E–07	6.59E–01
Np237	4.47E+07	1.92E–01	Tc 99	1.10E–05	8.55E–02	Cm245	1.94E–07	3.64E–01	Bi210	4.62E–07	5.32E–01
Sb126m	3.37E+07	1.45E–01	Po214	9.37E–06	7.26E–02	Am241	6.06E–08	1.14E–01	Zr 93	3.92E–07	4.51E–01
Sn126	3.37E+07	1.45E–01	Th230	7.32E–06	5.67E–02	Nb 93m	4.06E–08	7.62E–02	Cs135	3.55E–07	4.10E–01
Cs135	2.92E+07	1.25E–01	Po218	7.31E–06	5.67E–02	Pu242	3.66E–08	6.86E–02	Pb214	3.50E–07	4.03E–01
Sb126n	2.26E+07	9.68E–02	U-238	6.83E–06	5.29E–02	Pa234m	3.16E–08	5.92E–02	Np237	2.74E–07	3.16E–01
U-236	1.94E+07	8.32E–02	Rn222	6.68E–06	5.18E–02	U-234	2.53E–08	4.74E–02	Pu242	2.66E–07	3.06E–01
Am241	1.38E+07	5.90E–02	Po210	6.44E–06	4.99E–02	Th234	1.41E–08	2.64E–02	Sb126	2.55E–07	2.94E–01
Pu241	1.30E+07	5.55E–02	Ra226	5.82E–06	4.51E–02	Bi213	1.31E–08	2.46E–02	U-234	2.47E–07	2.84E–01
Cm245	1.29E+07	5.54E–02	Cm246	3.43E–06	2.66E–02	Th229	9.16E–09	1.72E–02	Cm245	1.68E–07	1.94E–01
Rest	1.48E+08	6.37E–01	Rest	1.84E–05	1.43E–01	Rest	6.04E–08	1.13E–01	Rest	7.44E–07	8.58E–01

3 Radiation dose rate to water

3.1 Alpha

The total alpha stopping power in H_2O and UO_2, and the alpha range in H_2O, has been calculated with the code ASTAR (Berger 1993), see Figure 3-1. The dominant alpha energies for the U-233 and BU60 pellets are from U-233 and Pu-240, respectively, see Table 2-3 and Table 2-6. The maximum alpha energies for these two nuclides are 4.9 and 5.2 MeV, respectively (Forrest 2007). The following conclusions may be drawn:

- The stopping power, expressed in MeV cm^2/g, is typically a factor 4 higher in H_2O compared to UO_2 in the considered energy interval 0 to 5 MeV.
- The maximum range for the alpha radiation in water for the considered energy interval 0 to 5 MeV is about 37 μm.

The alpha radiation dose rate outside the U-233 pellet has been calculated earlier (Jégou 2005). The results are summarized in Figure 3-2. The maximum alpha dose rate at the pellet surface is 680 Gy/h and the average alpha dose rate in the interval 0 to 34 μm is 82.8 Gy/h. The calculated alpha dose rate range in Figure 3-2, 34 μm, is in quite good agreement with the projected range for 4.9 MeV alpha particles presented in Figure 3-1.

The calculated maximum radiation at pellet surface, 680 Gy/h, can be roughly compared to the presented total alpha source term in Table 2-2:

- Total alpha dose rate inside UO_2 pellet: 111 Gy/h[4].
- Approximately 50 % in UO_2 at surface: 56 Gy/h.
- A factor 4 higher in water in contact with UO_2: 223 Gy/h.

Note that the above estimate is quite approximate and does not consider factors as density differences[5], etc. From that perspective the calculated dose rates in Jégou (2005) are considered to be in reasonable agreement with the calculated alpha source term.

The corresponding alpha dose rates for the BU60 pellet have been estimated based on the calculated results for the U-233 pellet, but corrected for the difference in alpha source strength. The results are presented in Table 3-1. The alpha dose rate for the BU60 pellet is slightly higher than the U-233 pellet at a decay time of 1 000 years, but slightly lower at decay periods of 3 000 and 10 000 years.

Table 3-1. Contact and average alpha dose rate on U-233 and BU60 pellet in water (1 000, 3 000 and 10 000 years).

	Alpha dose rate in water, Gy/h	
	Max at UO_2 surface	Average 0–34 μm
U-233 doped pellet	680	82,8
BU60 pellet, decay 1 000 y	1 111	135
BU60 pellet, decay 3 000 y	499	61
BU60 pellet, decay 10 000 y	281	34

4 0.031 W/kg × 3 600 s/h.

5 Density UO_2 10.5 g/cm^3, H_2O 1 g/cm^3.

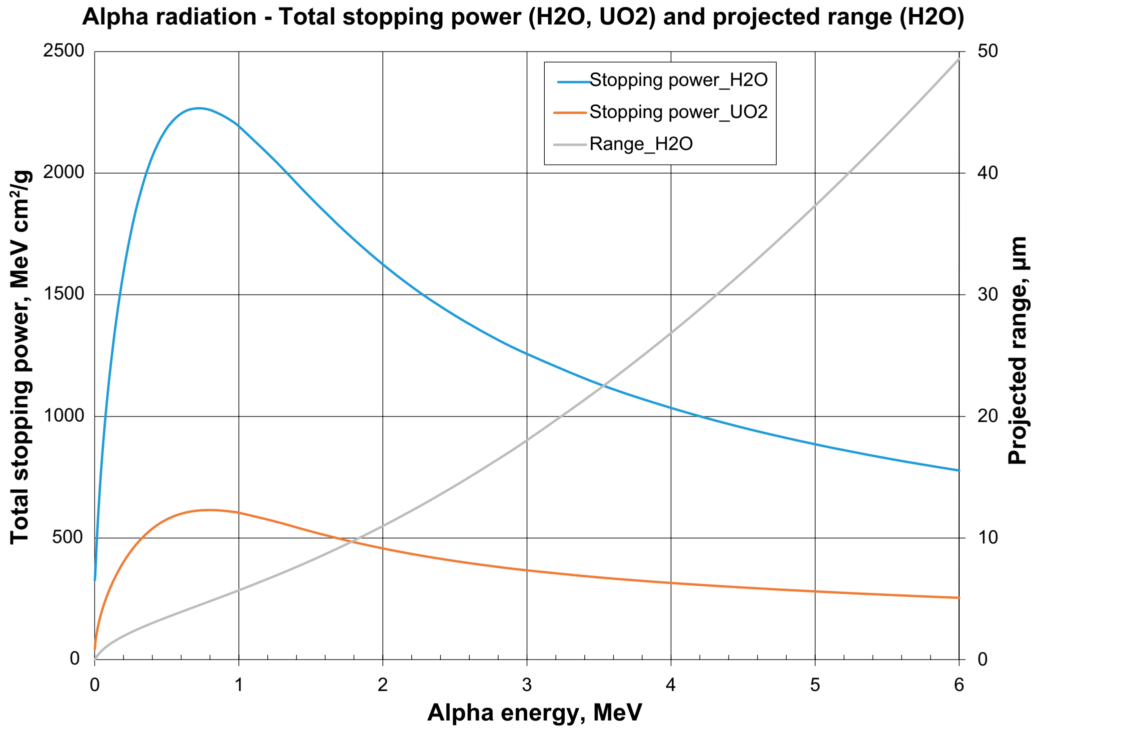

Figure 3-1. Alpha radiation – total stopping power in H_2O and UO_2 and projected range in H_2O based on computer code ASTAR (Berger 1993).

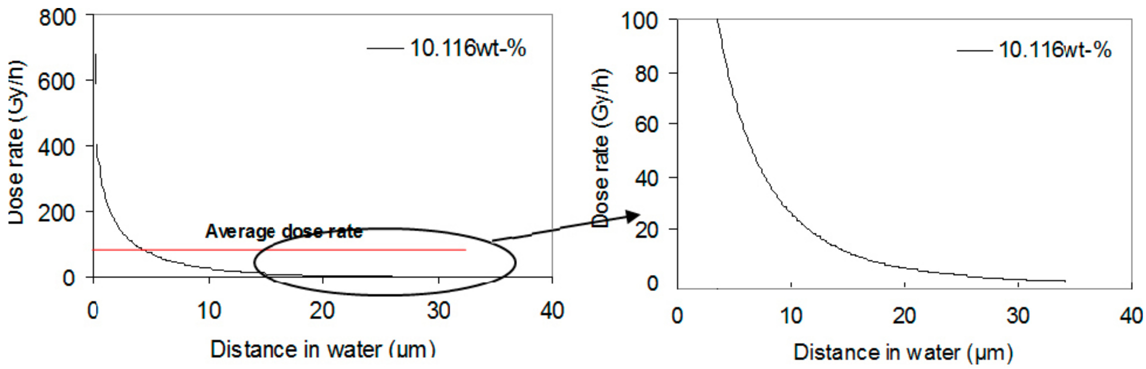

Figure 3-2. Calculated alpha dose rate in water outside U-233 doped fuel pellet (Jégou 2005).

3.2 Electrons

The total electron stopping power in H_2O and UO_2, and the range in H_2O, has been calculated with the code ESTAR (Berger 1993), see Figure 3-3. The electron energies for the U-233 and BU60 pellets for their dominant nuclides are shown in Figure 3-4 and Figure 3-5, respectively. The following conclusions may be drawn:

- The electron energies are at maximum about 2 MeV for the U-233 pellet (Bi-212 and Bi-213). The electron energies for the BU60 pellet are somewhat lower, <1 MeV.
- The maximum range for the electron radiation in water for the considered energy interval 0 to 2 MeV is about 10 000 µm. A realistic assumption is that the electron dose rate is quite constant in a water layer up to the alpha limit 34 µm, even considering electron energies << 1 MeV.
- The stopping power is typically a factor of 2 higher in water than in UO_2.

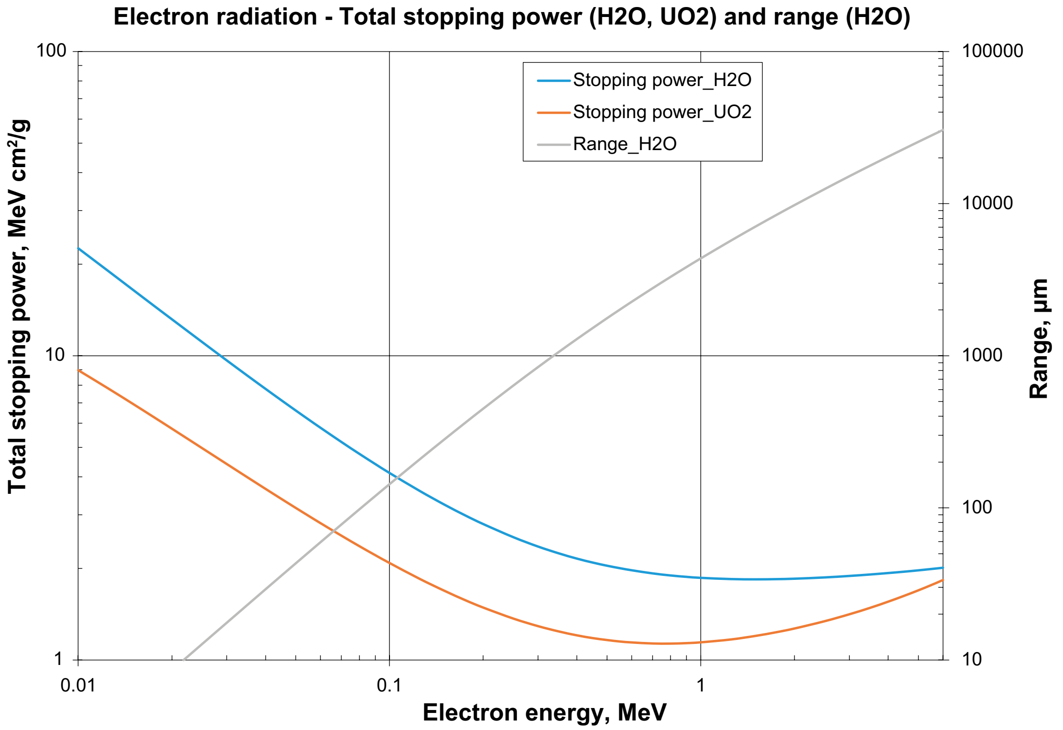

Figure 3-3. *Electron radiation – total stopping power in H_2O and UO_2 and range in H_2O based on computer code ESTAR (Berger 1993).*

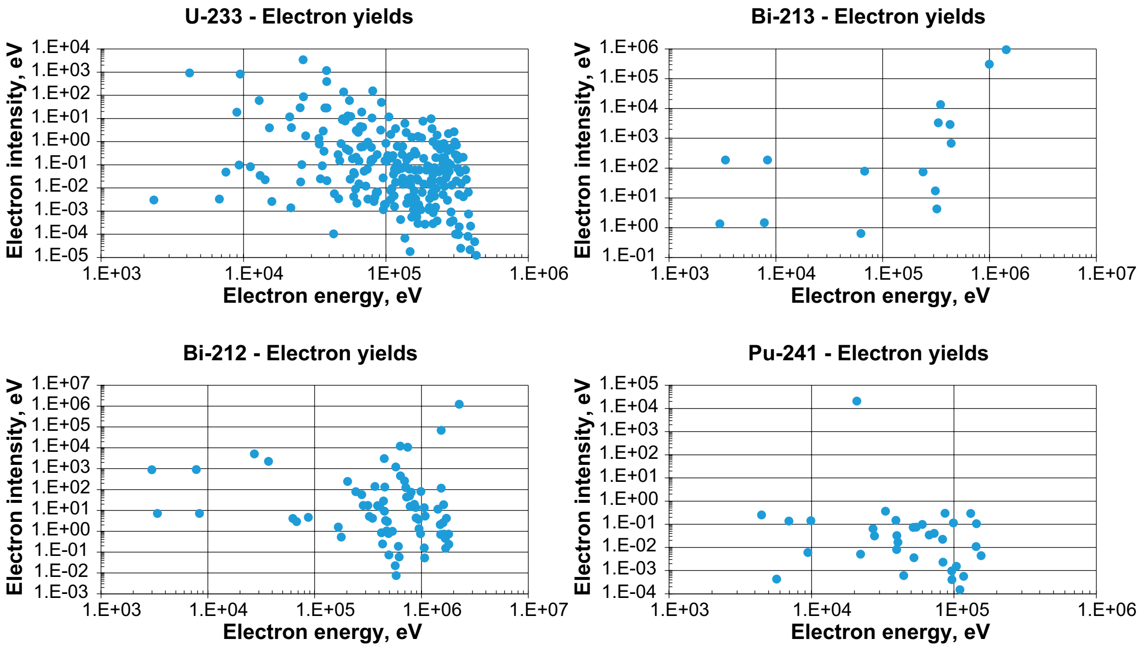

Figure 3-4. *U-233 pellet – electron yields for the four most dominant nuclides (Forrest 2007).*

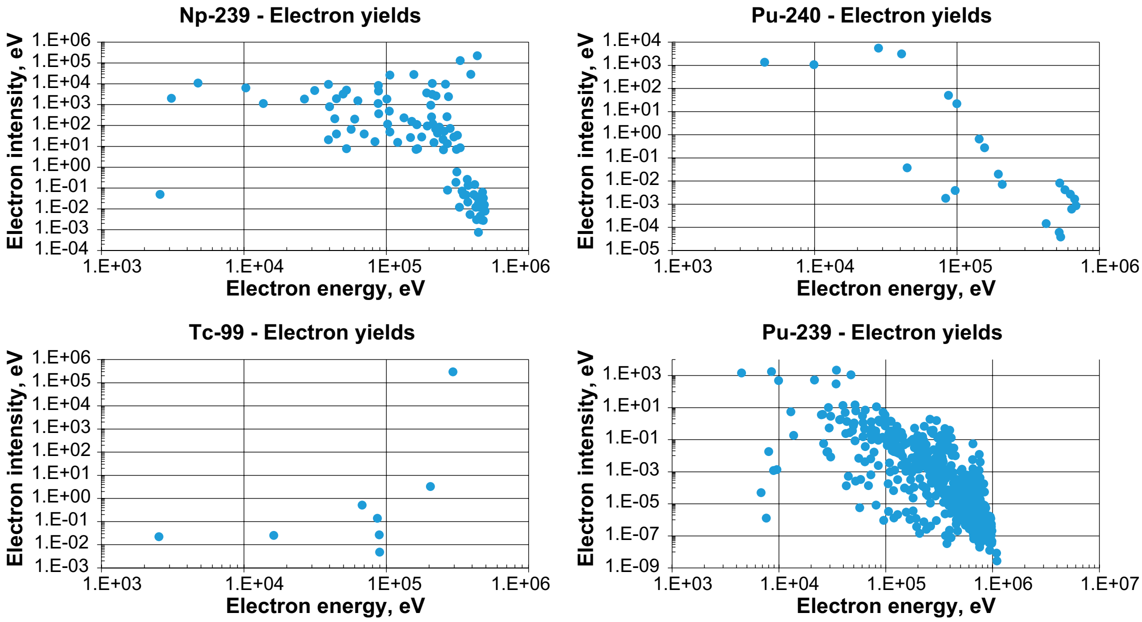

***Figure 3-5.** BU60 pellet (3 000 years) – electron yields for the four most dominant nuclides (Forrest 2007).*

The electron dose rates has been estimated in the following way:

- Electron source terms, W/kg UO_2, according to Table 2-3, Table 2-5, Table 2-6 and Table 2-7.
- Conversion to electron dose rate to water by considering surface effect (50 %) and higher stopping power in water (factor 2).

The resulting nuclide specific estimated electron dose rates to water in contact with the pellets are presented in Table 3-2. The electron dose rate outside the U-233 pellet is slightly lower than for the BU60 pellet for the decay periods 1 000, 3 000 and 10 000 years.

Table 3-2. Contact electron dose rate on U-233 and BU60 pellet in water (1 000, 3 000 and 10 000 years).

U-233_Pellet			BU60_Pellet, 1 000 years			BU60_Pellet, 3 000 years			BU60_Pellet, 10 000 years		
Nuclide	Dose rate	Percent	Nuclide	Dose rate	Percent	Nuclide	Dose rate	Percent	Nuclide	Dose rate	Percent
	(Gy/h)	Dose rate		(Gy/h)	Dose rate		(Gy/h)	Dose rate		(Gy/h)	Dose rate
Total	3.07E–01		Total	1.23E+00		Total	5.39E–01		Total	3.12E–01	
U-233	1.58E–01	5.13E+01	Am241	6.31E–01	5.12E+01	Np239	2.72E–01	5.05E+01	Np239	1.41E–01	4.50E+01
Bi213	3.29E–02	1.07E+01	Np239	3.28E–01	2.66E+01	Pu240	1.07E–01	1.98E+01	Pu240	5.10E–02	1.63E+01
Bi212	2.36E–02	7.67E+00	Pu240	1.32E–01	1.07E+01	Tc 99	4.06E–02	7.53E+00	Tc 99	3.97E–02	1.27E+01
Pu241	2.08E–02	6.77E+00	Tc 99	4.09E–02	3.32E+00	Pu239	3.16E–02	5.86E+00	Pu239	2.69E–02	8.61E+00
Pb209	1.46E–02	4.76E+00	Pu239	3.29E–02	2.67E+00	Am241	2.62E–02	4.86E+00	Am243	1.28E–02	4.10E+00
Tl208	9.85E–03	3.20E+00	Am243	2.99E–02	2.43E+00	Am243	2.48E–02	4.59E+00	Sb126m	1.22E–02	3.89E+00
Am241	9.69E–03	3.15E+00	Sb126m	1.25E–02	1.01E+00	Sb126m	1.24E–02	2.30E+00	Pa233	4.95E–03	1.59E+00
Th229	8.62E–03	2.81E+00	Pa234m	4.70E–03	3.81E–01	Pa233	4.93E–03	9.15E–01	Pa234m	4.70E–03	1.50E+00
Ra225	8.02E–03	2.61E+00	Pa233	4.33E–03	3.52E–01	Pa234m	4.70E–03	8.72E–01	Bi214	2.71E–03	8.66E–01
Pb212	7.91E–03	2.57E+00	Sn126	2.19E–03	1.77E–01	Sn126	2.17E–03	4.03E–01	Sn126	2.13E–03	6.81E–01
Pu238	3.36E–03	1.09E+00	Nb 93m	2.07E–03	1.68E–01	Nb 93m	2.06E–03	3.83E–01	Nb 93m	2.06E–03	6.59E–01
Ac225	2.05E–03	6.66E–01	Zr 93	1.42E–03	1.15E–01	Zr 93	1.41E–03	2.62E–01	Bi210	1.66E–03	5.32E–01
Pu239	1.90E–03	6.19E–01	Cs135	1.28E–03	1.04E–01	Cs135	1.28E–03	2.38E–01	Zr 93	1.41E–03	4.51E–01
Pu240	1.49E–03	4.84E–01	Cm245	1.26E–03	1.03E–01	Cm245	1.07E–03	1.99E–01	Cs135	1.28E–03	4.10E–01
Tl209	1.10E–03	3.57E–01	C 14	9.85E–04	7.99E–02	Np237	9.83E–04	1.82E–01	Pb214	1.26E–03	4.03E–01
Th228	9.59E–04	3.12E–01	Pu242	9.72E–04	7.89E–02	Pu242	9.69E–04	1.80E–01	Np237	9.87E–04	3.16E–01
U-232	7.67E–04	2.50E–01	Sb126	9.42E–04	7.65E–02	Sb126	9.37E–04	1.74E–01	Pu242	9.57E–04	3.06E–01
Fr221	6.54E–04	2.13E–01	U-234	9.09E–04	7.38E–02	U-234	9.05E–04	1.68E–01	Sb126	9.17E–04	2.94E–01
U-234	5.08E–04	1.65E–01	Np237	8.64E–04	7.01E–02	C 14	7.72E–04	1.43E–01	U-234	8.88E–04	2.84E–01
Pa234m	4.98E–04	1.62E–01	Pu238	6.24E–04	5.06E–02	Bi214	4.61E–04	8.54E–02	Cm245	6.06E–04	1.94E–01
Rest	2.77E–04	9.01E–02	Rest	2.26E–03	1.83E–01	Rest	2.11E–03	3.91E–01	Rest	2.68E–03	8.58E–01

3.3 Gamma

The FISPACT-II calculations, besides radioactivity and total source terms, also yield gamma spectra and gamma contact dose rates[6], considering the attenuation properties in the UO_2 material.

The gamma source spectra in the U-233 and BU60 pellets are presented in Figure 3-6.

The calculated gamma contact dose rates for the U-233 doped and BU60 pellets are presented in Table 3-3. The gamma dose rate for the U-233 pellet is slightly higher than the corresponding for the BU60 pellet, even for the shortest decay period 1 000 years. The gamma dose rate should be very constant in a water layer up to the alpha limit 34 µm.

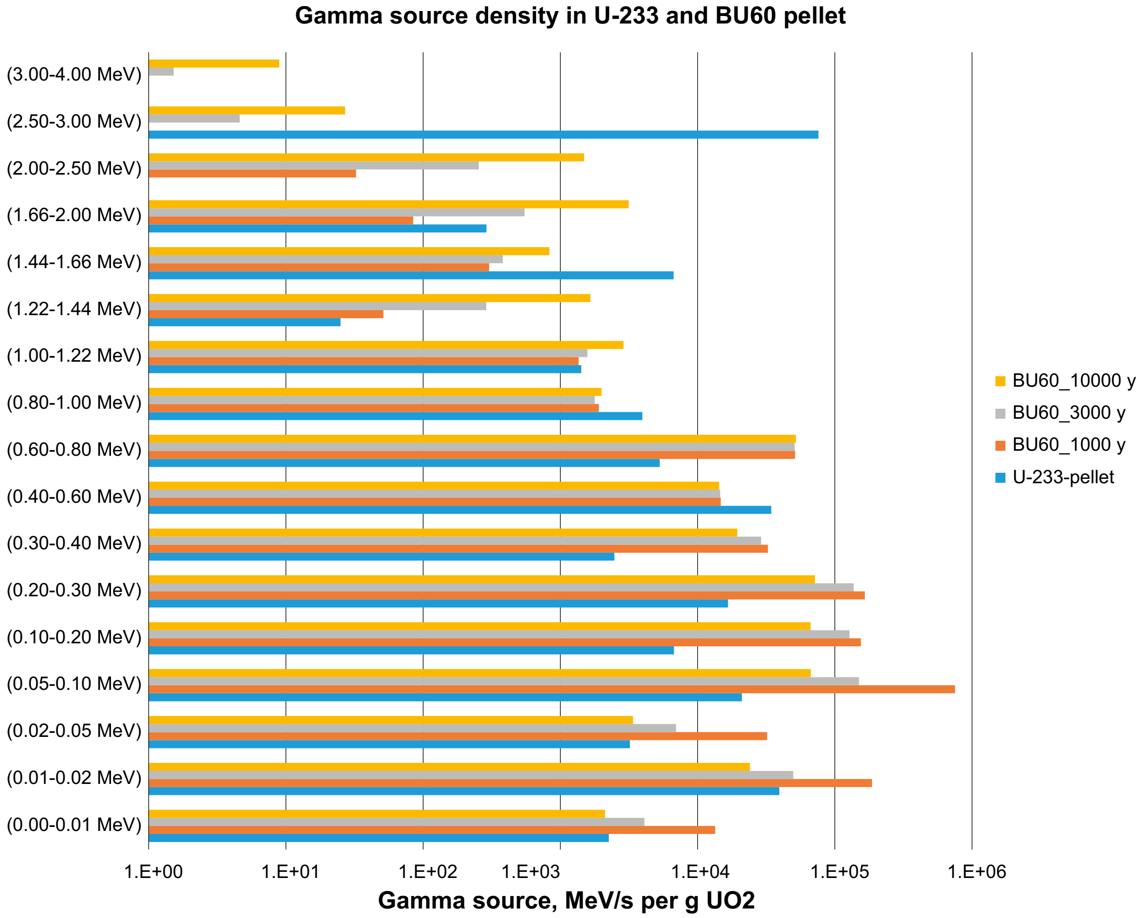

Figure 3-6. *Comparison gamma source spectra in U-233 and BU60 pellets.*

[6] The calculated gamma contact dose rate is for a semi-infinite slab with the prescribed material composition.

Table 3-3. Contact gamma dose rate on U-233 and BU60 pellet in water (1 000, 3 000 and 10 000 years).

U-233_Pellet			BU60_Pellet, 1 000 years			BU60_Pellet, 3 000 years			BU60_Pellet, 10 000 years		
Nuclide	**Dose rate**	**Percent**	**Nuclide**	**Dose rate**	**Percent**	**Nuclide**	**Dose rate**	**Percent**	**Nuclide**	**Dose rate**	**Percent**
	(Gy/h)	**Dose rate**		**(Gy/h)**	**Dose rate**		**(Gy/h)**	**Dose rate**		**(Gy/h)**	**Dose rate**
Total	2.72E–02		Total	2.41E–02		Total	1.79E–02		Total	1.67E–02	
Tl208	2.19E–02	8.05E+01	Sb126m	7.98E–03	3.32E+01	Sb126m	7.93E–03	4.43E+01	Sb126m	7.77E–03	4.65E+01
Bi212	1.49E–03	5.48E+00	Np239	6.84E–03	2.84E+01	Np239	5.67E–03	3.17E+01	Np239	2.93E–03	1.76E+01
Bi213	1.48E–03	5.46E+00	Am241	5.37E–03	2.23E+01	Sb126	2.03E–03	1.13E+01	Bi214	2.88E–03	1.72E+01
Tl209	1.20E–03	4.40E+00	Sb126	2.04E–03	8.48E+00	Am243	5.68E–04	3.17E+00	Sb126	1.99E–03	1.19E+01
U-233	5.95E–04	2.19E+00	Am243	6.86E–04	2.85E+00	Bi214	4.90E–04	2.74E+00	Pa233	4.27E–04	2.56E+00
Pb212	2.26E–04	8.30E–01	Pu240	4.18E–04	1.74E+00	Pa233	4.26E–04	2.38E+00	Am243	2.94E–04	1.76E+00
Th229	8.61E–05	3.17E–01	Pa233	3.74E–04	1.55E+00	Pu240	3.38E–04	1.89E+00	Pu240	1.62E–04	9.67E–01
Fr221	7.91E–05	2.91E–01	Ho166m	1.01E–04	4.20E–01	Am241	2.23E–04	1.25E+00	Pb214	8.39E–05	5.02E–01
Am241	7.15E–05	2.63E–01	Pu239	9.18E–05	3.82E–01	Pu239	8.81E–05	4.92E–01	Pu239	7.51E–05	4.49E–01
Ac225	2.57E–05	9.48E–02	Bi214	6.25E–05	2.60E–01	Pa234m	3.58E–05	2.00E–01	Pa234m	3.58E–05	2.14E–01
Ra224	1.76E–05	6.48E–02	Pa234m	3.58E–05	1.49E–01	Ho166m	3.18E–05	1.78E–01	Np237	1.09E–05	6.54E–02
Pu238	1.04E–05	3.83E–02	Cm245	1.78E–05	7.40E–02	Cm245	1.51E–05	8.45E–02	Sn126	9.37E–06	5.61E–02
Ra225	7.00E–06	2.58E–02	Sn126	9.63E–06	4.00E–02	Pb214	1.43E–05	7.98E–02	Cm245	8.55E–06	5.12E–02
Pu239	4.59E–06	1.69E–02	Np237	9.56E–06	3.98E–02	Np237	1.09E–05	6.08E–02	Bi213	8.44E–06	5.05E–02
Rn220	4.35E–06	1.60E–02	Nb 93m	3.81E–06	1.59E–02	Sn126	9.57E–06	5.35E–02	Tl209	6.75E–06	4.04E–02
Pu240	4.08E–06	1.50E–02	Pa234	3.81E–06	1.59E–02	Pa234	3.81E–06	2.13E–02	Pa234	3.81E–06	2.28E–02
Pa234m	3.31E–06	1.22E–02	Pu242	3.35E–06	1.39E–02	Nb 93m	3.81E–06	2.13E–02	Nb 93m	3.80E–06	2.27E–02
Th228	2.68E–06	9.86E–03	Np238	2.33E–06	9.69E–03	Pu242	3.34E–06	1.87E–02	Pu242	3.30E–06	1.97E–02
At217	2.05E–06	7.55E–03	U-234	2.32E–06	9.63E–03	U-234	2.31E–06	1.29E–02	Am241	2.64E–06	1.58E–02
U-232	1.62E–06	5.98E–03	Pu238	2.23E–06	9.27E–03	Nb 94	2.05E–06	1.15E–02	U-234	2.27E–06	1.36E–02
Rest	2.79E–06	1.03E–02	Rest	7.35E–06	3.06E–02	Rest	4.26E–06	2.38E–02	Rest	8.52E–06	5.10E–02

4 Discussion and summary

A summary of the presented dose rates to water outside the U-233 doped and BU60 pellets is presented in Table 4-1. Some comments and conclusions:

- The average alpha dose rate in a water layer up to 34 μm is about a factor of 8 lower than the alpha dose rate in contact with the pellet.
- The average electron and gamma dose rates are estimated to be quite constant within the alpha 0 to 34 μm range.
- The average alpha dose rates are significantly higher than the electron dose rates (a factor of 110 to 270) and the gamma dose rates (a factor of 2 000 to 5 600)[7].
- All dose rates for the U-233 doped pellet are quite similar to the dose rates for the BU60 pellet at the considered decay periods of 1 000, 3 000 and 10 000 years.
  - The electron dose rate for the alpha-doped pellet is ~ 0.4 % of the average alpha dose rate, while in the case of the 3 000 years old fuel it is ~ 0.9 % of the average alpha dose rate. The gamma dose rate is ~ 0.033 % of the average alpha dose rate for the alpha-doped pellet and ~ 0.030 % for the 3 000 years old fuel.

Table 4-1. U-233 and BU60 pellet (1 000, 3 000 and 1 000 years) – Dose rate to water in contact – Summary.

	Alpha dose rate in water, Gy/h		Electron dose rate in water, Gy/h		Gamma dose rate in water, Gy/h	
	Max at UO_2 surface	Average 0–34 μm	Max at UO_2 surface	Average 0–34 μm	Max at UO_2 surface	Average 0–34 μm
U-233 doped pellet	680	82.8	0.31	0.31	0.027	0.027
BU60 pellet, decay 1 000 y	1 111	135	1.23	1.23	0.024	0.024
BU60 pellet, decay 3 000 y	499	61	0.54	0.54	0.018	0.018
BU60 pellet, decay 10 000 y	281	34	0.31	0.31	0.017	0.017

The above results may be compared to the corresponding results for CANDU fuel with burnup 7.9 MWd/kgU (Sunder 1998). The following approximate results were obtained for a decay period of 3 000 years:

- Alpha: = 20 Gy/h (average 0–35 μm).
- Electron: = 0.01 Gy/h (average 0–400 μm).
- Gamma: = 0.001 Gy/h.
  - It is concluded in Sunder (1998) that the gamma dose rate is significantly higher, typically a factor of 6, if surrounding fuel is considered.

The differences between the BU60 and the CANDU results are reasonable considering very different burnup levels, different water layers for electrons, etc. The CANDU results, however, support the overall conclusion that the dose rate to water is very much dominated by the alpha contribution at a decay period of about 3 000 years.

[7] Note that the radiolysis yields for the alpha high LET radiation may be quite different compared to the yields from low LET electron and gamma radiation.

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