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The degradation of gamma-ray mass attenuation of UO₂ and MOX fuel with nuclear burnup

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Abstract

Nondestructive gamma-ray spectrometry of nuclear fuel is routinely performed in axial gamma scanning devices and more recently with gamma emission tomography. Following the irradiation of a fresh nuclear fuel with high intensity neutron flux in a nuclear reactor core, a great number of gamma-emitting radionuclides are created. These can be utilized for gamma spectrometric techniques. However, due to the high density and atomic number of the nuclear fuel, self-attenuation of gamma-rays is a challenge, which requires attenuation correction in order to perform accurate analysis of the source activity in the fuel.

In this study, the degradation of the gamma-ray mass attenuation with burnup was investigated and, in addition, a predictive model was created by investigating the attenuation change at various gamma energies caused by the burnup of the nuclear fuel. This model is intended for use by spectrometry practitioners inspecting nuclear fuel. To this aim, the energy-dependent gamma-ray mass-attenuation coefficients were investigated as a function of burnup for UO₂, and three MOX fuels having different initial Pu contents. The Serpent 2 reactor physics code was used to simulate the burnup history of the fuel pins. The nuclide inventory of the Serpent 2 output is combined with the NIST XCOM database to calculate the mass attenuation coefficients.

The mass attenuation coefficient of the fuel was found to decrease with the fuel burnup, in the range of a few percent, depending on the burnup and gamma energy. Also, a theoretical burnup dependent swelling model was imposed on fuel density to see how linear attenuation coefficient of fuel material is changed. Furthermore, greater effect may be expected on the transmitted intensity, where a simulation study of a PWR assembly revealed that the contribution from the inner rods in a scanned fuel assembly increased by tens of percent compared to the one with non-irradiated fresh fuels, when shielded by the outer rods of the assembly. A sensitivity analysis was performed in order to test the effect of a number of geometrical and operational reactor parameters that were considered to potentially effect the mass attenuation coefficient. Finally, a simple-to-use predictive model was constructed providing the mass-attenuation coefficient [cm²/g] of fuel as a function of burnup

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[MWd/kg_{HM}] and initial Pu content [wt%]. The resulting predictive model was optimized by using the nonlinear regression method.

Keywords: Gamma-ray spectrometry; emission tomography; nuclear fuel assembly; nondestructive measurements; high burnup; gamma-ray self-attenuation.

1. Introduction

Gamma-ray spectrometry (henceforth spectrometry) is a method widely used for nondestructive inspections of nuclear fuel. While fresh nuclear fuel is only very weakly radioactive, the operation of the fuel in the artificial neutron flux of nuclear reactor causes the additional creation of an abundance of artificial radionuclides, many of which are gamma-ray emitters.

Spectrometry of nuclear fuel assemblies is performed in axial gamma scanning devices (1), as well as more recently in gamma emission tomography (2), (3), (4). The spectrometric technique has many nuclear-fuel related applications, including:

- Determination of burnup and power distributions of the nuclear fuel rod for code validation (5), (6).
- Process monitoring and verification of burnup history in reprocessing plants (7), (8).
- Determination of fission gas release (9), (10).
- Characterization of molten core material after nuclear accidents (11), (12)
- Identification and quantification of relocated of nuclear fuels in LOCA transient tests (13)
- In the nuclear safeguards field, for detection of partial defects or missing fuel pins (14) or determination of fissile content (15).

In the techniques listed above, the gamma-rays emitted from nuclear fuel are collimated and then measured by a spectroscopic detector. The gamma-ray field is collimated to limit the sensitivity of the system to a selected part of the nuclear fuel, and the system may be moved relative to the fuel in order to provide information on the spatial distributions of selected radionuclides. Some examples of commonly used radionuclides for nuclear fuel spectrometry are given in Table 1. As can be seen, there is a wide range of gamma energies available for assessment.

Table 1. Typical gamma-emitting nuclides in used nuclear fuel (a non-exclusive list, both in terms of nuclides and in terms of their gamma-ray energies). Nuclides in parenthesis are a short-lived decay product that emits the gamma energy listed (the daughter may often be in secular equilibrium with the mother nuclide).

Nuclide	Half-life	Gamma energy [keV]	Creation mode
Cs-137	30.1 y	661.7	Fission product
Eu-154	8.6 y	123.1, 1274.4	Activation of Eu-153
Cs-134	2.1 y	604.7, 795.9	Activation of Cs-133
Co-60	5.3 y	1173.3, 1332.5	Activation of Co-59
Ce-144 (Pr-144)	284 d (17 min)	2185.7	Fission product
Zr-95	64 d	724.2, 756.7	Fission product

Nb-95	35 d	765.8	Fission product and decay of Zr-95
Ba-140 (La-140)	13 d (1.7 d)	1596.2	Fission product
I-131	8 d	364.5	Fission product

An ever-present challenge in achieving accurate quantitative spectrometry is to properly account for the self-attenuation of the sample under investigation. This problem is especially challenging when performing spectrometry on nuclear fuel, which presents a very high attenuation of the gamma-ray intensity due to its combination of high density and high-Z composition.

In principle, accounting for the self-attenuation in nuclear fuel would be a trivial task, since the as-manufactured dimensions and material constitution of the fuel are well-known; however, the task becomes more complicated when the fuel undergoes nuclear burnup. The replacement of each fissioned atom by approximately two lighter fission products leads to changes in the elemental composition and fuel dimensions that differ increasingly from the as-manufactured specifications as burnup increases. It can be noted that in many of the applications of spectrometry on nuclear fuels listed in section 1, the fuel may be assessed after the end-of-life in the reactor. Thereby, the fuel typically has burnup that locally reaches above 50 MWd/kg_{HM}, approximately equivalent to 5% of the initial uranium atoms being consumed, and even higher burnup is possible.

The presented investigations in this article aimed to investigate the evolution of the mass-attenuation coefficient in the most common LWR nuclear fuel (UOX and MOX), in order to answer the question if the burnup causes important changes. In addition, to create a predictive model that can be used to correct for such attenuation changes, in order to achieve accurate estimates of the self-attenuation in spectrometric assay of nuclear fuel.

2. Theory

The gamma-rays from nuclear fuel rod are self-attenuated in the fuel due to gamma-ray interactions with the fuel material, and in the case of inspections of a fuel assembly, the gamma-rays are also attenuated by fuel rods in front of the neighboring rods. For accurate analysis of the source activity using gamma spectrometry, the self-attenuation needs to be accounted for when calculating the source activity from the measured peak intensity. In the case of tomography, it equivalently needs to be accounted for in the calculation of the system matrix (16). One method for this purpose is the analytical correction according to Eq. 1, where I is the transmitted intensity, I_0 is the unattenuated intensity, x is the material thickness, ρ is the density and μ is the linear attenuation coefficient of the material. For convenience, the ratio of μ/ρ , the mass attenuation coefficient, is often used.

$$I/I_0 = \exp[-(\mu/\rho)x\rho] \quad (1)$$

Obtaining an accurate mass attenuation coefficient is thus crucial for the correction of the self-attenuation of gamma-rays in the fuel. This coefficient varies with the elemental composition of the fuel and the gamma energy. The gamma-rays of interest to nondestructive assay of nuclear fuel are typically in the range from 100 to 2000 keV. In this range, the gamma-ray interaction modes are photoelectric absorption, Compton scattering, and pair production.

In the photoelectric absorption process, the gamma-ray loses all of its energy in one interaction. The probability for this process depends strongly on gamma-ray energy and atomic number of the material, and approximately proportional to Z^n/E^m , where Z is the atomic number and E is the energy of the gamma-ray, and n and m are exponents varying from 3-5 (17). Therefore, the photoelectric absorption reaction is dominating in the low gamma energy range and for high- Z materials. In Compton scattering, the gamma-ray loses only part of its energy in one interaction. Still, this is enough to consider the gamma-ray to be lost from the perspective of spectrometry, where the full energy peak is normally used. The probability for the Compton process is weakly dependent on E and Z . Finally, the pair-production interaction, which is possible only for gamma energies above 1.022 MeV, also has a cross section that increases in importance with Z .

Clearly, considering the high- Z of uranium, the interaction modes that depend strongly on Z (photoelectric absorption and pair production) are important for large sections of the gamma energy range. Therefore, the hypothesis behind this work was that the fission of heavy nuclides in the fuel of high burnup can cause a non-negligible decrease of the mass attenuation coefficient of gamma-rays. Figure 1 shows the evolution of the concentration of fission products, uranium and other actinides with the burnup. While it can be noted that the main trend is the gradual replacement of heavy uranium with lighter fission products, which decreases the effective Z of the material and reduces the attenuation, there is also a competing phenomenon of the breeding of transuranic actinides. Thereby, the inclusion of these high- Z elements increases the attenuation of the gamma-rays in the nuclear fuel.

Figure 1. Evolution of Uranium concentration, fission products and actinides with burnup. The uranium content was excluded from the actinide graph.

In addition to the changing elemental composition of nuclear fuel due to burnup, it is also well known that fuel swelling occurs (18), (19), (20). In accordance with eq. 1, changes in the density, ρ , will also affect the transmission of gamma-rays through the fuel. However, as stated in Section 3.1, in the context of our investigations, this effect was excluded in the simulations, and instead we refer to earlier work on the models for nuclear fuel swelling with burnup. According to reference (19) the volumetric swelling is 0.95 % per 10 MWd/kg_{HM} up to 57 MWd/kg_{HM}, and at greater burnup, it decreases to 0.32% per 10 MWd/kg_{HM}.

3. Methods and Materials

3.1 Serpent 2:

In this work, the elemental composition of the used nuclear fuel was obtained by calculation with Serpent 2. This is a multi-purpose three-dimensional continuous-energy Monte Carlo particle transport code, developed at VTT Technical Research Centre of Finland, Ltd. The radioactive decay constant and fission yield data are read from ENDF/B-VI.8 and, as neutron cross section library, JEFF-3.1 is used. Serpent can be run as a completely stand-alone application without dependence to external depletion codes or pre-generated data sets (21).

In this work, an infinite two-dimensional lattice of pin-cells fueled by both UO₂ and MOX fuels was modelled and simulated by the Serpent 2.1.28 code. Table 2 shows the geometrical modelling parameters.

Table 2. Geometrical parameters used to model infinite two-dimensional lattice system of pin-cells in Serpent 2.

Parameter	Value
Fuel radius	4.10 mm
Clad inner radius	4.18 mm
Clad outer radius	4.75 mm
Pitch	16 mm

Liquid water was modelled as coolant and moderator, with density and temperature of 0.74 g/cm³ and 600 K respectively. The cladding was purely Zirconium, and cladding and fuel temperatures were set at 600 K and 900 K, respectively. The Uranium was enriched to 4.9 wt% in U-235 in UO₂ fuel and three levels of initial Pu mass content were selected for MOX fuels, which were 2.9%, 4.4%, and 5.6% respectively. The burnup steps were chosen to be 2 MWd/kg_{HM}, which corresponds to an irradiation time of about 49 days, in order to obtain a detailed evolution of the isotopic composition, and the reactor was simulated to run for a total burnup of up to 110 MWd/kg_{HM}. This high burnup value was chosen to include a burnup range that, with the potential improvements in fuel performance, could possibly be reached in the near future, and to include the burnup of the fuel rods of the Halden LOCA test series, which occasionally reached above 90 MWd/kg_{HM} (22). Also, some simplifications are made throughout the simulations such as homogeneous burnup within the fuel, no density and structural changes associated with swelling or densification caused by burnup. Furthermore, no migration of fission products is modelled and its effect was found to be negligible as given in Section 9. All simulation parameters are listed in Table 3.

Table 3. Simulation parameters used to model infinite two-dimensional lattice system of pin-cells in Serpent 2.

Parameter	Value
Burnup	110 MWd/kg _{HM}
Linear heat	200 W/cm
Water temperature	600 K
Fuel temperature	900 K
Clad temperature	600 K

3.2 NIST database

In this work, the mass attenuation coefficient of the individual elements, μ/ρ , were obtained from the NIST (National Institute of Standards and Technology) XCOM database (23). NIST XCOM contains energy-dependent mass-attenuation coefficients, μ/ρ , for all elements $Z = 1$ to 100, including all absorption edges. The available μ/ρ values are tabulated according to theoretical values of the total cross section per atom, σ_{tot} , which relates to μ/ρ as,

$$\mu/\rho = \sigma_{tot}/(uA) \quad (2)$$

where u is the atomic mass unit, A is the relative atomic mass of the target element, and σ_{tot} is the total cross section with coherent scattering for an interaction by the photon given in units of barns/atom where $1 \text{ barn} = 10^{-24} \text{ cm}^2$. As indicated before, the attenuation coefficient, photon interaction cross sections and related quantities are functions of the photon energy. The total cross section is the sum of the contributions from the principal photon interactions,

$$\mu/\rho = (\sigma_{pe} + \sigma_{comp} + \sigma_{pair})/(uA) \quad (3)$$

where σ_{pe} is the atomic photoelectric cross section, σ_{comp} is the Compton scattering cross section, and σ_{pair} is the pair production cross section.

For chemical compositions and mixtures, the following summation rule of Eq. 4 is applied, where w_i is the fraction by weight of atomic constituent i obtained from the Serpent 2 depletion code, and the $(\mu/\rho)_i$ values are from the NIST XCOM database.

$$\mu/\rho = \sum_i w_i (\mu/\rho)_i \quad (4)$$

3.3 Postprocessing of Serpent2 output

All numerical output of Serpent, by default, is written in MATLAB® m-files to simplify the post-processing of the results. The Serpent output includes the mass fractions of each isotope/element in the fuel up to any atomic number selected by the user (in this work $Z=98$). The Serpent output gives this for each burnup step of 2 MWd/kg_{HM}. In all simulations, data that is lost to undefined nuclides were found negligible. The nuclide weight fractions are

provided with the Serpent output. For the energy dependent mass attenuation values, log-log interpolation was performed between the listed energies in NIST XCOM.

3.4 Uncertainty considerations:

For the statistical uncertainty, each test problem was simulated many times with different random seed. For a test case of UO_2 fuel representing our simulations, the relative statistical error of k_{eff} was found to be nearly 0.035%, while mass densities of U and Pu elements had a relative statistical error of about 0.00062% and 0.034%, respectively. By using error propagation, the relative uncertainty (1σ) of the mass attenuation coefficient calculated at a burnup of 110 MWd/kg_{HM} was found to be 3 ppm at the energy of 123.1 keV. This is considered to be negligible, and the error bars were omitted from the figures. Consequently, the uncertainty can be expected to be dominated by systematic error sources, due to e.g. inaccuracy of the geometrical model of the reactor and the fuel. The importance of the variation of such parameters is described in more detail in a sensitivity analysis of Section 5.

4. Results

Figure 2 presents the evolution of the mass attenuation coefficient in UO_2 fuel for three selected gamma-ray energies that were selected to represent a low, an intermediate and a high gamma energy used in spectroscopic measurements, 123.1, 661.7 and 1596.2 keV. The values in the graphs were normalized by the fresh fuel case, in order to facilitate interpretation of the evolution with burnup. It can be seen that the attenuation is decreasing with the burnup. This was expected since the effective atomic number (Z) of the fuel material is decreasing during burnup making the photoelectric effect less effective as defined in Section 2. The change in attenuation can be several percent and it is more pronounced towards lower energies and higher burnup due to the same reasons. For the standard gamma energy used in spectrometry of nuclear fuel of the 661.7 keV peak of ^{137}Cs , the reduction of the mass-attenuation is between 3% - 5% at the burnup interval of 60 - 110 MWd/kg_{HM}.

Figure 2. Evolution of mass attenuation coefficient with burnup for the selected three gamma-ray energies in UO_2 fuel.

As for the MOX fuels, the change of mass attenuation coefficient with burnup was investigated for three initial Pu contents completely the same way with that of UO_2 fuel. The results were found to be nearly the same as in the case of UO_2 fuel as can be deduced from Figure 3 for all the three simulations, where attenuation changes between about 3% -5% at the burnup interval of 60 – 110 $\text{MWd/kg}_{\text{HM}}$ for the 661.7 keV peak of ^{137}Cs . Again, for the reasons given above, it is more pronounced at the lower energies. Taking into account the change of mass attenuation with burnup in UO_2 and MOX fuels as given in Figures 2 and 3, a clear overview of the gamma-ray energy regions where the change is more important may be shown as in Figure 4 for three selected burnup values: 30, 70, and 110 $\text{MWd/kg}_{\text{HM}}$. The change in low energy regions is due to dominant photoelectric effect as stated above. As can be seen, at higher than 1500 keV the absolute value of the percentage change starts to slightly increase. This is due to Z dependence of pair production cross section as in low energy regions, which was expressed in Section 2.

Figure 3. Evolution of mass attenuation coefficient with burnup for the selected three gamma-ray energies in MOX fuel with different Pu content.

Figure 4. Percent change in mass attenuation coefficient with energy at the three selected burnup values in UO_2 fuel.

The evolution of the mass attenuation coefficients with burnup is shown in Figure 5 for all simulated types of fuels at the selected low gamma-ray energy value, 123.1 keV. At the burnup level of 110 MWd/kg_{HM}, the difference between the calculated mass attenuation coefficients of UO₂ and MOX with 5.6% Pu fuels is calculated as 4.44e-3, which corresponds to a negligible relative difference of 0.13%. This difference is mainly due to initial U and Pu contents of the fuels. It was deduced from the graphs that one could establish a relationship that predicts the fuel mass attenuation coefficients as a function of burnup and initial Pu content. Thus, the generated data was fitted to a 2D plane equation at various gamma-ray energies such that the mass attenuation coefficient was expressed as,

$$\mu/\rho = a_0 + a_1BU + a_2Pu \quad (5)$$

where BU and Pu are the burnup [MWd/kg_{HM}] and initial Plutonium content [%] of the MOX fuel respectively, and a_0 , a_1 and a_2 are the fitted coefficients. The model coefficients were fitted using least squares regression with the MATLAB® function `nlinfit`, to the Serpent2-based calculated mass-attenuation coefficients.

Figure 5. The evolution of the mass attenuation coefficients with burnup for all simulated types of fuels for the selected low gamma-ray energy of 123.1 keV.

The coefficients of Eq. 5 have been listed for a wide range of gamma energies in Table 4 along with their 95% confidence bounds. In addition to the regular energy interval, values are also given explicitly at the discontinuities at the K-edges, to facilitate interpolation in their vicinity.

Table 4. Coefficients of the predictive model of Eq. 5. It can be noted that for the UO_2 fuel, where the initial Pu-content is zero, the coefficient a_2 can be ignored. (Element in parentheses of column one are the parent element of the K-edge, where applicable)

E (keV)	a_0	a_1	a_2
100	1.7424 (1.7421 1.7425)	-0.000118 (-0.000120 -0.000116)	0.000731 (0.000698 0.000764)
101.1 (Fr-87)	1.6960 (1.6958 1.6961)	-0.000117 (-0.000118 -0.000115)	0.000711 (0.000678 0.000742)
103.9 (Ra-88)	1.5896 (1.5894 1.5897)	-0.000113 (-0.000114 -0.000111)	0.000663 (0.000633 0.000693)
106.7 (Ac-89)	1.4908 (1.4906 1.4909)	-0.000110 (-0.000111 -0.000108)	0.000619 (0.000591 0.000647)
109.6 (Th-90)	1.3987 (1.3986 1.3988)	-0.000106 (-0.000107 -0.000104)	0.000579 (0.000552 0.000604)
110	1.3882 (1.3880 1.3883)	-0.000106 (-0.000107 -0.000104)	0.000574 (0.000548 0.000600)
112.5 (Pa-91)	1.3131 (1.3129 1.3132)	-0.000102 (-0.000103 -0.000101)	0.000541 (0.000516 0.000565)
115.6 (U-92)	4.2896 (4.2866 4.2926)	-0.002883 (-0.002917 -0.002847)	-0.012498 (-0.013119 -0.011875)
118.6 (Np-93)	4.0233 (4.020 4.0261)	-0.002697 (-0.002729 -0.002663)	-0.011805 (-0.012387 -0.011223)
120	3.9182 (3.9154 3.9209)	-0.002627 (-0.002658 -0.002594)	-0.011495 (-0.012061 -0.010927)
121.8 (Pu-94)	3.8204 (3.8200 3.8206)	-0.003018 (-0.003021 -0.003014)	0.000733 (0.000668 0.000796)
125.0 (Am-95)	3.5860 (3.5857 3.5862)	-0.002785 (-0.002787 -0.002782)	0.001024 (0.00097 0.001072)
128.2 (Cm-96)	3.3705 (3.3703 3.3707)	-0.002544 (-0.002545 -0.002541)	0.001084 (0.001040 0.001127)
130	3.2583 (3.2580 3.2584)	-0.002459 (-0.002461 -0.002456)	0.001051 (0.001008 0.001093)
131.6 (Bk-97)	3.1628 (3.1626 3.1630)	-0.002387 (-0.002389 -0.002385)	0.001022 (0.000981 0.001063)
136.0 (Cf-98)	2.9200 (2.919 2.9201)	-0.002205 (-0.002206 -0.002202)	0.000949 (0.000910 0.000987)
140	2.7215 (2.7213 2.7216)	-0.002055 (-0.002057 -0.002053)	0.000889 (0.000853 0.000925)
160	1.9733 (1.9731 1.9733)	-0.001483 (-0.001484 -0.001481)	0.000657 (0.000630 0.000684)
180	1.4902 (1.4901 1.4902)	-0.001109 (-0.001110 -0.001107)	0.000503 (0.000482 0.000523)
200	1.1598 (1.1597 1.1598)	-0.000854 (-0.000855 -0.000853)	0.000396 (0.000379 0.000411)
250	0.7051 (0.7051 0.7051)	-0.000493 (-0.000494 -0.000492)	0.000238 (0.000228 0.000247)
300	0.4707 (0.4707 0.4707)	-0.000313 (-0.000314 -0.000312)	0.000157 (0.000150 0.000163)
400	0.2691 (0.2691 0.2691)	-0.000156 (-0.000157 -0.000155)	8.40e-05 (8.07e-05 8.77e-05)
500	0.1847 (0.1846 0.1846)	-9.20e-05 (-9.21e-05 -9.14e-05)	5.30e-05 (5.10e-05 5.55e-05)
600	0.1410 (0.1409 0.1410)	-6.00e-05 (-6.02e-05 -5.97e-05)	3.70e-05 (3.54e-05 3.85e-05)
800	0.0980 (0.0980 0.0980)	-3.10e-05 (-3.12e-05 -3.07e-05)	2.20e-05 (2.09e-05 2.28e-05)
1000	0.0772 (0.0771 0.0771)	-1.80e-05 (-1.84e-05 -1.78e-05)	1.50e-05 (1.40e-05 1.53e-05)
1250	0.0629 (0.0629 0.0629)	-1.10e-05 (-1.12e-05 -1.08e-05)	1.00e-05 (9.95e-06 1.08e-05)
1500	0.0554 (0.0554 0.0554)	-8.00e-06 (-8.24e-06 -7.98e-06)	8.00e-06 (7.94e-06 8.78e-06)
2000	0.0483 (0.0483 0.0483)	-7.00e-06 (-7.12e-06 -6.89e-06)	7.00e-06 (6.49e-06 7.08e-06)
2500	0.0456 (0.0455 0.0455)	-7.00e-06 (-7.17e-06 -6.86e-06)	6.00e-06 (5.77e-06 6.29e-06)

5. Sensitivity analysis

Sensitivity analysis of the results were performed in order to examine the impact of various geometrical and operational parameters of the fuel and the coolant/moderator in the reactor core on the calculated mass attenuation coefficients. For this purpose, a large number of individual simulations were performed by changing only one operational parameter from its nominal value. The operational parameters that were varied are the moderator density, fuel enrichment, soluble boron concentration, linear power, and fuel and cladding temperatures. The soluble boron concentrations were kept constant over the whole burnup. A combined effect of the operational parameters such as Gd_2O_3 together with borated water was also investigated, in addition to the separate effects mentioned above. All these parameters and the range they were changed are summarized in Table 5 at a gamma-ray energy of 123.1 keV. In addition, the pitch of the fuel assembly was varied in order to test the sensitivity to the geometry, e.g. affecting the moderator to fuel ratio. Furthermore, the maximum relative change of μ/ρ from the nominal conditions is shown.

Table 5. Parameters varied in the sensitivity analysis together with the relative change of μ/ρ in comparisons to nominal conditions at a gamma-ray energy of 123.1 keV. It can be noted that the imposed variations in the analysis are much larger than what may be expected in the normal operation of nuclear fuel in the cases of moderator density, linear power and the pitch size.

Parameter	Range	Max. change (ppm)
Moderator density	0.04 – 0.74 g/cm ³	2
Fuel enrichment	Natural U – 20 wt%	0.6
Soluble boron	No boron – 650 ppm	0.6
Linear power	50– 500 W/cm	<0.01
Fuel & clad temperature	900 – 1500 K	0.5
Pitch	12– 28 mm	1.0

Simulation results of sensitivity analysis have shown a relative change of about 2 ppm with respect to the mass attenuation coefficients calculated at nominal conditions even at higher burnups, e.g. at 110 MWd/kg_{HM}. In general, it may be concluded that our predictive model is reasonably robust across different parameter sets.

6. Case Study 1: Calculation of the linear attenuation coefficient

Indeed, it is the total attenuation change that the spectrometry practitioner will need finally. In this part, we have calculated the change in gamma-ray linear attenuation coefficients with burnup for the typical 661.7 keV of ¹³⁷Cs according to Eq.1. This has been performed in three different ways. First, we have only taken into account the change in mass attenuation. Then, by keeping mass attenuation unchanged with burnup, we did only include the effect of density

change due to a simple swelling model given in (19). Finally, both effects were combined to evaluate the linear attenuation coefficient. Resulting attenuation values are shown in Figure 6 as normalized to fresh fuel to simplify the analysis.

Figure 6. Evolution of linear attenuation coefficient with burnup in UO_2 fuel for 661.7 keV of ^{137}Cs for the cases: change in only mass attenuation, only density, and combined effect.

For a gamma-ray transmitted through 100 MWd/kg_{HM} fuel, the swelling is about 6.8 % according to (19), causing a reduction of the linear attenuation coefficient of about 6% as can be seen in Fig.6. This can be compared to the reduction due to the mass-attenuation change studied by us, which is energy dependent, of about 4 %. In this case, the combined effect is nearly 10%. These decreases of the gamma-ray attenuation cause additive errors if not accounted for. However, accounting for the swelling is less straightforward, since the reduction of density is accompanied by increase of the volume, and thus an increase of the path-length of the gamma-rays traversed in the fuel. This should be analyzed with more sophisticated fuel performance codes coupled with fuel burnup codes, which may be regarded as a future work.

7. Case Study 2: Applications to gamma scanning

In references (24) and (25), a measurement system used for axial and rotational scanning while performing gamma spectroscopy assay of nuclear fuel assemblies is presented. A model

of the measurement system, with a 17×17 PWR fuel assembly, was created in Geant4. The model was used to study the flux of gamma radiation of 662 keV from ^{137}Cs from various fuel rods in the assembly, through the opening of collimator away from the fuel. Figure 7 displays a simplified geometry of the model, indicating the coordinate system used for the calculations. Two calculations were performed; a) The flux with a fuel pellet material consisting of fresh, non-irradiated, UO_2 . b) The flux with a fuel pellet material composed of the material simulated using the procedure described in section 3, for a burnup of 48 $\text{MWd/kg}_{\text{HM}}$ and an initial enrichment of 5%. The results of the two calculations are presented in Table 6. As can be seen, with increasing amount of material, the change in self-attenuation and the resulting photon flux increasingly more and more significant.

Figure 7. A simplified view of the geometry used in model of the gamma scanning system. The PWR 17×17 fuel assembly (F) was rotated 45 degree with a corner facing the collimator slit (S). The distance between the centre of the fuel assembly and the detector (D), including the wall (W) was about 2.5 meter. The coordinate axes (x,y) indicate the position of rods inside the fuel assembly. The detector (D) indicate which side of the collimator open where the gamma-ray flux was calculated.

Table 6. The flux of 662 keV gamma radiation through the gamma scanning collimator slit, with fresh fuel pellet material (column “fresh flux”) and with a fuel pellet material corresponding to a burnup of 48 $\text{MWd/kg}_{\text{HM}}$ and initial enrichment of 5% (column “irradiated flux”). Fuel rods located on the diagonal of the fuel assembly (with the diagonal corner pointing towards the detector) were used here.

Fuel rod position (x,y) [cm]	Fresh flux [1/cm ² /emitted particle] × 10 ¹¹	Irradiated flux [1/cm ² /emitted particle] × 10 ¹¹	Relative difference [%]
(10.08,-10.08)	610(4)	606(8)	0.72
(8.82,-8.82)	236(3)	236(5)	0.05
(7.56,-7.56)	92(2)	96(3)	-4.1
(5.04,-5.04)	33(1)	36(2)	-8.7

(2.52,-2.52)	10.9(5)	14(2)	-20
(1.26,-1.26)	5.3(4)	6(1)	-11
(-1.26,1.26)	2.1(3)	2.6(7)	-21

8. Conclusions

The gamma spectroscopy technique is widely used for characterization of the nuclear fuel. In this technique, the activity of specific nuclides is measured by the quantification of the count-rates of their characteristic gamma-rays in the detector. However, the composition of the nuclear fuel changes with burnup due to nuclear transmutation, which impacts the self-attenuation of the gamma-rays. This needs to be accounted for in order to obtain accurate results of the spectrometric assay. In this paper, we investigated the variation of the mass attenuation coefficients of UO₂ and MOX fuels with the burnup and we concluded to the following:

- 1) It was found that the mass attenuation coefficient of the nuclear fuel material decreases with burnup especially at lower gamma-ray energies due to the dominant photoelectric effect. This was in accordance with our expectations, since the atomic number of the constituent elements decreases with fission. The magnitude of the change was inspected for a wide range of gamma energies. For the commonly used 661.7 keV peak of ¹³⁷Cs it was found to decrease between 3% - 5% at the high burnup interval of 60 - 110 MWd/kg_{HM}.
- 2) To provide a simple correction for the change of the attenuation, a predictive model has been established for the burnup-dependent mass attenuation coefficients of the UOX and MOX fuels, using the gamma-ray energy, the burnup and the initial plutonium content (in case of MOX) as input parameters. The linear model was fitted using least squares regression to the calculated mass attenuation coefficients based on Serpent 2 simulations and the NIST XCOM elemental attenuation data.
- 3) The effect on the evolution of the mass-attenuation coefficient of other reactor or fuel parameters, such as moderator density, fuel enrichment, soluble boron concentration, linear power, or physical dimensions, were all found to be negligible. The effect of these parameters on the mass attenuation coefficient of nuclear fuel was of a smaller order of magnitude than the effect of the burnup.
- 4) Statistical analysis show a small relative uncertainty in the nuclide densities obtained from the burnup simulations. Propagation of these uncertainties to the calculated mass attenuation coefficients revealed that the statistical uncertainty source is of a negligible order of magnitude.

In the light of our findings above, we propose the usage of the burnup-corrected mass attenuation coefficients of UOX and MOX fuels obtained in eq. 5 for gamma spectrometry measurements on nuclear fuel. The coefficients, a_1 - a_3 , are interpolated with respect to the gamma energy, using the simple look-up table of table 4.

9. Discussion

A major simplification in this work was the assumption of homogeneous burnup distribution within the fuel. In a real nuclear fuel there are burnup gradients across various length-scales, within the reactor core e.g. due to neutron leakage at the perimeter and due to control rods, within the assemblies. Variations can be expected also due to the neutronic rim effect, inducing a higher fission rate in the periphery of the rods, where pellet-rim burnup could be approximately 1.8-2 times higher than the pellet average (26). In principle, the burnup profile can be calculated based on operator data and the local burnup may in that case be used with eq. 5. to achieve the corresponding attenuation coefficient.

In addition to the decrease of the mass attenuation coefficient investigated in this paper, fuel swelling or densification associated with burnup affects the density evolution of the fuel, which in turn, according to eq. 1, also affects the transmitted gamma-ray intensity. However, there is prior work by others that have resulted in models for fuel swelling, e.g. (20), (19), that may be applied by spectrometry practitioners to correct for the density evolution of the nuclear fuel. Although this was outside the scope of our work, we made simplified analysis to show the importance of density change. In addition to swelling, the higher burnup that is achieved at the rim area is significant, and structural changes have been observed there (27), which can be expected to cause a density inhomogeneity in the fuel. However, the impact of such features on spectrometry of the fuel has not been considered in our models.

Finally, it can be noted that fission gasses are produced as a result of the fission product yield of noble gasses, primarily xenon and krypton. These gasses may be dissolved in the fuel grains, trapped in intragranular bubbles, or in bubbles on the grain face or edges (28). In these cases, they will contribute to the mass attenuation coefficient such as modelled in our work. However, fission gasses may also be released from the fuel to the gap and plenum of the fuel rod, for example caused by power transients (28), in which case it is lost from the fuel and should not be taken into account in the mass attenuation. In the calculations of the mass attenuation coefficient in our work, we have assumed that all fission gasses remained trapped in the fuel. At the gamma-ray energy of 121.3 keV, summed impact of the primary noble gasses to the total mass attenuation was found to be about 0.6% at the burnup of 110 MWd/kg_{HM}, which means an insignificant contribution.

10. Outlook

As noted in the discussion above, the fuel may undergo structural changes, swelling or densification, and fission product migration due to the burnup in a nuclear reactor. These phenomena may impact local attenuation of gamma-rays. This may be accounted for by the use of coupled fuel depletion and fuel performance codes, e.g. SEN (Serpent and Enigma) (29), in order to obtain radial variations of burnup and also to include structural changes of the fuel, which finally may provide a more complete estimation of the density change of nuclear fuel with burnup. In addition, since the predictive model achieved in this work is based on reactor physics simulation results, it is proposed that an experimental validation is performed.

Outside the field of gamma-ray spectrometry, the gamma-ray attenuation decrease in nuclear fuel with burnup may also be of importance for the accurate calculation the canister surface temperature in final disposals. It can be noted that the decrease of the mass attenuation coefficient with burnup will cause a lower heat generation in the fuel, and relocate that heat production to the surrounding medium in the spent nuclear fuel, thereby affecting the temperature gradient in the ambient of the disposed fuel.

As a future work, we are planning to investigate the degradation of the gamma-ray attenuation in also Fast Reactor (FR) fuel in order to be used by the spectroscopy practitioners doing research in Gen-IV reactors. Compared with the UOX fuel of Light Water Reactors FR fuels may have higher burnup values, higher actinide concentration and swelling, which is considered to have an enhanced degrading effect on attenuation.

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