NON-DESTRUCTIVE FISSION GAS RELEASE DETERMINATION OF IRRADIATED EXPERIMENTAL FUEL RODS USING GAMMA SPECTROMETRY

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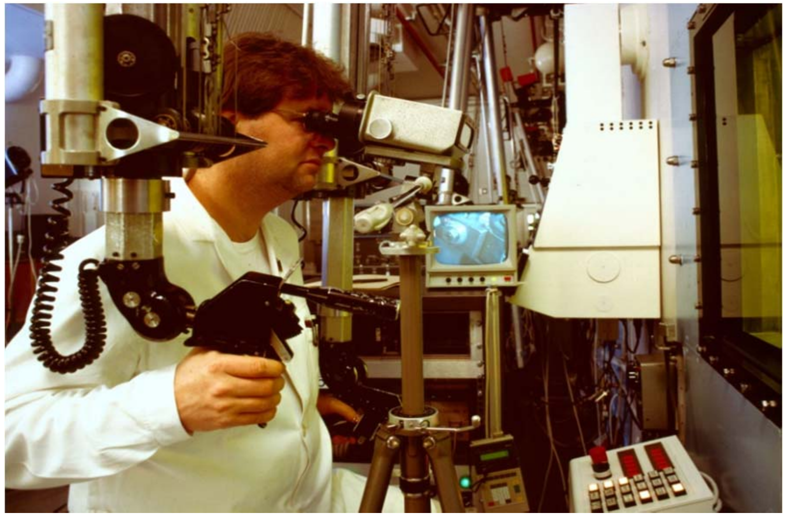


Overview

- Kr-85 method 1
- Kr-85-method 2
- Cs-137 and Kr-85 production and decay calculations
- Interaction of gamma radiation with matter
- Calculation and simulation of gamma escape propabilities
- Efficiency calculations of gamma radiation
- Examples

Kr-85 method 1

- Assuming the release rate of fission gas is equal for all gas atoms and isotopes, the number of Kr-85 gas atom in the plenum is given by,
- $N_{Kr85} = F_{tot} \times \gamma_{Kr85} \times FGR \times D_{Kr85},$
- D_{Kr85} is the effect of decay and γ_{Kr85} is the fission yield of Kr-85
- When the number of detected Kr-85 in the plenum by gamma spectrometry is R_{Kr85}, a correlation between R_{Kr85} and N_{Kr85} is as follows,
- $R_{Kr85} \times V_{free-volume} = N_{Kr85} \times \lambda_{Kr85} \times A_{Kr85} \times B_{Kr85} \times E_{Kr85}$
- $V_{free\text{-}volume}$ is the free volume of the fuel rod and λ_{Kr85} is the decay constant of Kr-85. A_{Kr85} is the γ-ray abundance (emission probability through disintegration of γ ray emission) of Kr-85 which is defined as number of photon emitted per decay and its value is 0.434%. B_{Kr85} is the effect of absorption, which means a ratio of Kr-85 escaping from absorption by plenum spring, cladding (escape probability). E_{Kr85} is the detector and setup efficiency.
- $\Leftrightarrow F_{tot} = R_{Kr85} \times \frac{V_{free-volume}}{\gamma_{Kr85} \times D_{Kr85} \times \lambda_{Kr85} \times A_{Kr85} \times B_{Kr85} \times E_{Kr85}} \times \frac{1}{FGR}$

Kr-85 method 1

- Following the same way as for Kr-85, the number of detected Cs-137 in the fuel stack R_{Cs137} is given by,
- $R_{Cs137} \times V_{fuel} = F_{tot} \times \gamma_{Cs137} \times D_{Cs137} \times \lambda_{Cs137} \times A_{Cs137} \times B_{Cs137} \times E_{Cs137}$
- $\Leftrightarrow F_{tot} = R_{Cs137} \times \frac{V_{fuel}}{\gamma_{Cs137} \times D_{Cs137} \times \lambda_{Cs137} \times A_{Cs137} \times B_{Cs137} \times E_{Cs137}}$
- Relating the two expressions for F_{tot} we get,
- $FGR = \frac{R_{Kr85} \times V_{free-volume} \times \gamma_{Cs137} \times D_{Cs137} \times \lambda_{Cs137} \times A_{Cs137} \times B_{Cs137} \times E_{Cs137}}{R_{Cs137} \times V_{fuel} \times \gamma_{Kr85} \times D_{Kr85} \times \lambda_{Kr85} \times A_{Kr85} \times B_{Kr85} \times E_{Kr85}}$

Kr-85 method 2

- $n_i = \frac{C_i^{decay} \times C_i^{shielding}}{E_{i,Kr85} \times VA_{Kr85} \times A_{Kr85}} \times I_i$
- n_i equals the volume of Kr-85 per unit length at STP conditions of fuel rod i at the end of irradiation (EOI), i.e. [cm^3/mm]. I_i is the γ-ray peak intensity (cps) of Kr-85 of the fuel rod i. C_i^{decay} is the compensation coefficient of the decay of Kr-85 of the fuel rod i from the end of irradiation (EOI) to the day of measurements. A_{Kr85} is defined earlier.
- $C_i^{decay} = e^{\frac{ln2}{T_{0.5}}(t_{spec} - t_{EOI})}$
- where $T_{0.5}$ is the half-life period of Kr-85 (10.78 years) and (t_{spec}-t_{EOI}) is the time period from end of irradiation to the day of gamma scanning.
- $C_i^{shielding}$ (B_{Kr85i}^{-1}) is the compensation coefficient (escape propability^{-1}) of the γ-ray shielding of the fuel rod i and E_i is the detector efficiency of Kr-85 at 514 keV $[\frac{cps}{Bq/mm_{plenum}}]$. VA_{Kr85} is the Kr-85 intensity per volume at STP, i.e. 0.55×10^{11} Bq/cm^3.

Kr-85 method 2

- $V({}^{85}Kr_{i,EOI}^{released}) = n_i \times \frac{V_i^{free}}{S_i}$ i.e. gas emission volume (cm^3 of Kr-85 at STP) of the fuel rod i at EOI. S_i is the cross section area of the fuel rod (interior) for fuel rod i. V_i^{free} is the free-volume of the fuel rod i at the EOI.
- When calculating the total gas volume from the volume of Kr-85, the Kr-85/Kr ratio as well as the Xe/Kr ratio are necessary to know. The experiences from earlier chemical analysis of fission gasses are used for estimation of these parameters.
- The total gas volume of [Xe and Kr] at STP is calculated using the following equation,
- $V(Xe + Kr)_{i,EOI}^{released} = [1 + (Xe/Kr)_{i,EOI}] \times \frac{V({}^{85}Kr_{i,EOI}^{released})}{({}^{85}Kr/Kr)_{i,EOI}}$
- $V(Xe + Kr)_{i,EOI}^{released}$ is equal to gas emission volume at EOI of the fuel rod i and $(Xe/Kr)_{i,EOI}$ is the Xe/Kr ratio at EOI of the fuel rod i and $({}^{85}Kr/Kr)_{i,EOI}$ is the Kr-85/Kr ratio at EOI of the fuel rod i.

Kr-85 method 2

- The fission gas release (FGR) is expressed as the total fission gas released to the total fission gas produced in the fuel.
- $FGR_i = \frac{(Xe+Kr)_{i,EOI}^{released}}{(Xe+Kr)_{i,EOI}^{produced}} \times 100\ [\%]$

Production and decay calculations

- The general solution for the irradiation / decay problem includes solution of the following differential equation,
- $\frac{dN_i}{dt} = \gamma_i \times F - \lambda_i \times N_i$
- The index i represent the isotope. N is the isotope density and λ is the disintegration constant. γ is the fission yield and F is the fission rate. If we assume the fission yield and rate to be constant, the following solution gives the isotope density during the j'th irradiation and decay cycle
- $N_{j,i}(t) = \frac{\gamma_i F_j}{\lambda_i}\left(1 - e^{-\lambda_i t}\right) + N_{j-1,i} e^{-\lambda_i t}$
- This expression must be used several time when many irradiation cycles are performed and t equals the irradiation time or cooling time when only decay takes place. $N_{j-1,\ i}$ equals the isotope density just before the j'th irradiation and decay cycle starts, i.e. $N_{j,i}(0) = N_{j-1,i}$. The index j represents the various irradiation and decay cycle. The total number of produces atoms is $F_j \times T_{irradiation}$. This number is used for normalisation of the irradiation/decay result $N_{j,i}(t)$.

Gamma ray absorption with matter

- Calculations of absorption effects (escape probabilities) for gamma rays traversing the fuel sample itself (self-absorption) and other shielding materials (lead) utilized in the setup for Kr-85 measurements is performed with “microShield” radiation software from Grove.
- This software can handle many sources and shields configurations and thereby making the calculation possible in an easy way. Cylindrical source and clad with various lead shields is the configuration normally utilized in the simulations performed for Kr-85 measurements of fuel rods. The calculation results (i.e. exposure mR/hr) are compared to similar results obtained from transmission of the actual gamma rays in air. The end results from the calculations give the escape probabilities for the gamma rays, i.e. the fraction reaching the detector area.
- MicroShield simplifies the amount of work relative to the normal way of manual calculations involving the modified Bessel and Struve functions
- The calculations are used to eliminate geometrical features, fuel and clad diameter.

Efficiency calculations

- For utilization of method 1, it is only necessary to know the relative efficiency (E_{Cs137}/E_{Kr85}) between the gamma signals of cs-137 (661.66 keV) in the fuel stack and for Kr-85 (514 keV) in the plenum. This can be achieved by acquisition of gamma spectrum from the actual fuel rod itself. By relating the cs-134 peak signals in the fuel spectrum, it is possible to estimate the linear relative efficiencies for the fission gas release estimations. It's no need for any other calibration sources. The definition for the setup efficiency is given by
- $E_i = \frac{I_i}{Activity_i \times B_i \times A_i}$
- Where the index i represents the different energy peaks belonging to the specific source isotopes. The B_i is calculated by using the radiation software from Grove.
- By comparing the fractions of various energy peaks in a specific isotope the activity is the same and is thereby not needed. However, for absolute efficiency calculations, a calibration fuel rod is suitable.

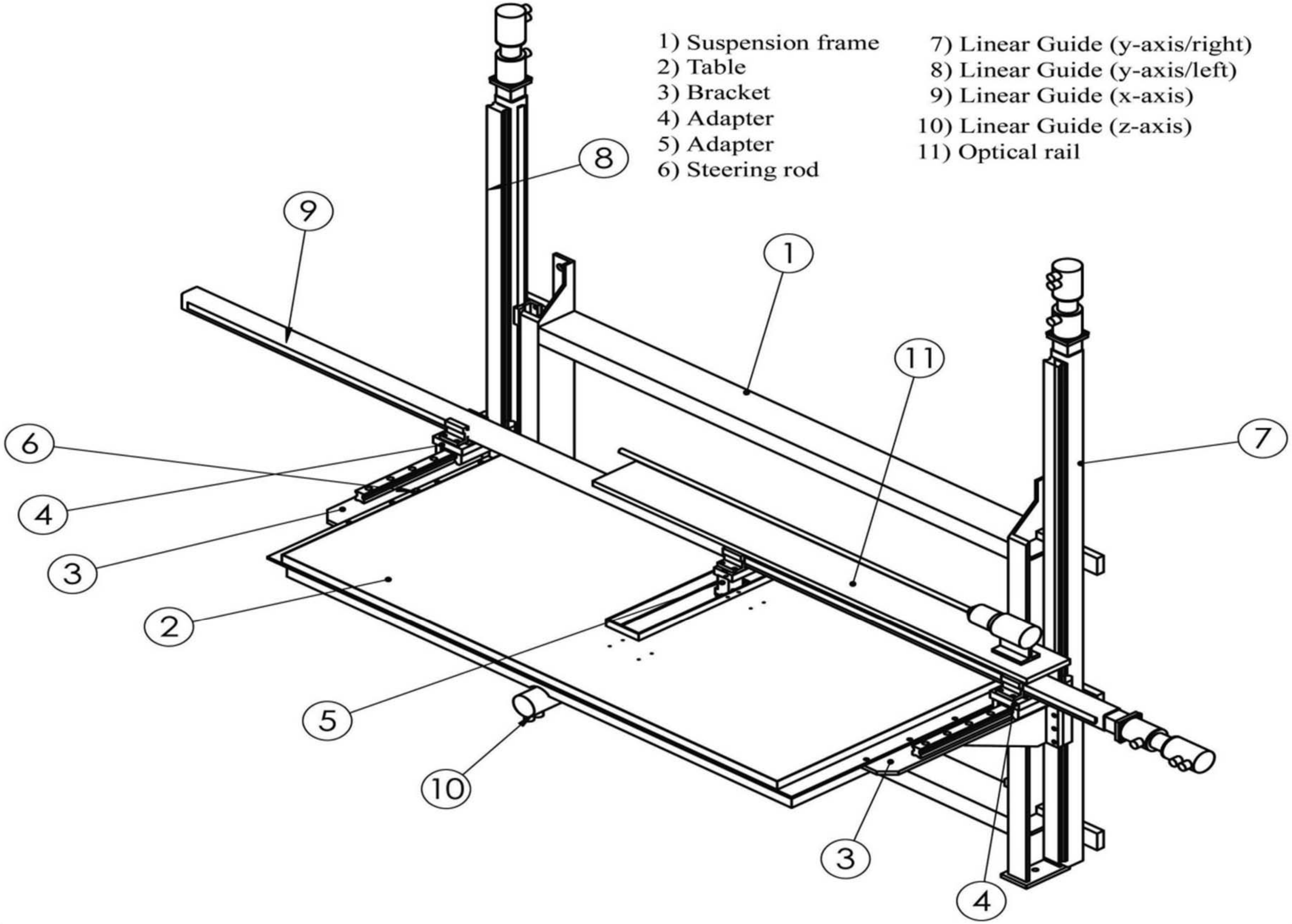
Efficiency calculations

- Cs-137 peak activity is first measured and utilized in the absolute efficiency calculations. The assumption of a linear efficiency behavior is a good approximation in the energy area (514 keV to 795 keV) of interests.
- The accuracy in the results will increase with utilization of relatively long acquisition times for the gamma spectra (e.g. 2-5 days) and for larger cs-134 activity in the fuel rod.

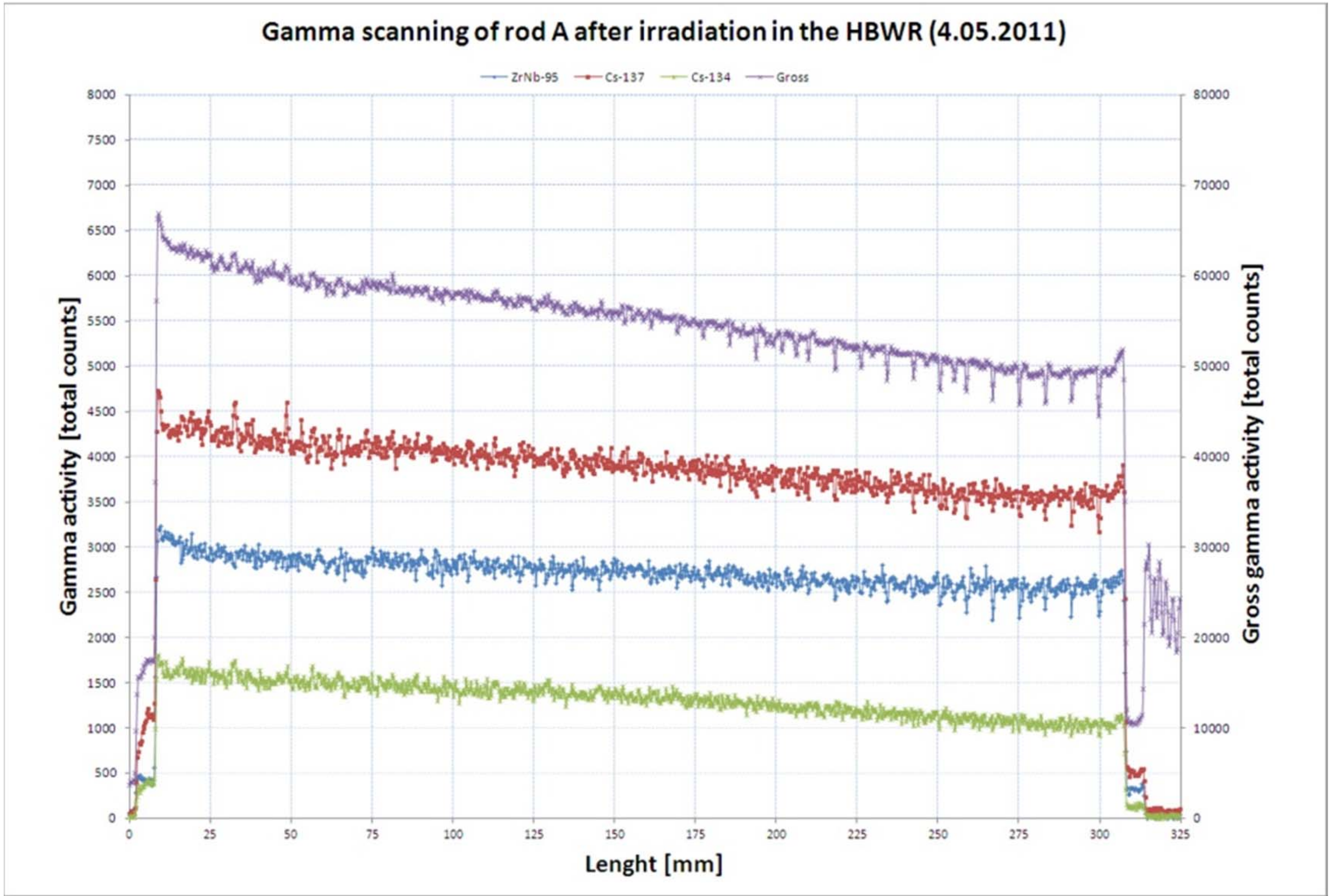
Test fuel rod and collimation and bench details for Kr-85 and cesium measurements

- UO_2 fuel rod with 16% U-235 enrichment irradiated in 4 cycles up to 58 MWd/kgUO_2 and 5.5 years irradiation time.
- 8.36 mm and 9.5 mm fuel and clad diameter respectively.
- 1.307 cm^3 free volume and 15 cm^3 fuel volume.
- Collimator 0.3 mm width and 8 mm lead shielding slab in front of the detector.
- 3-axis gamma scanning bench with rotation posibilities.

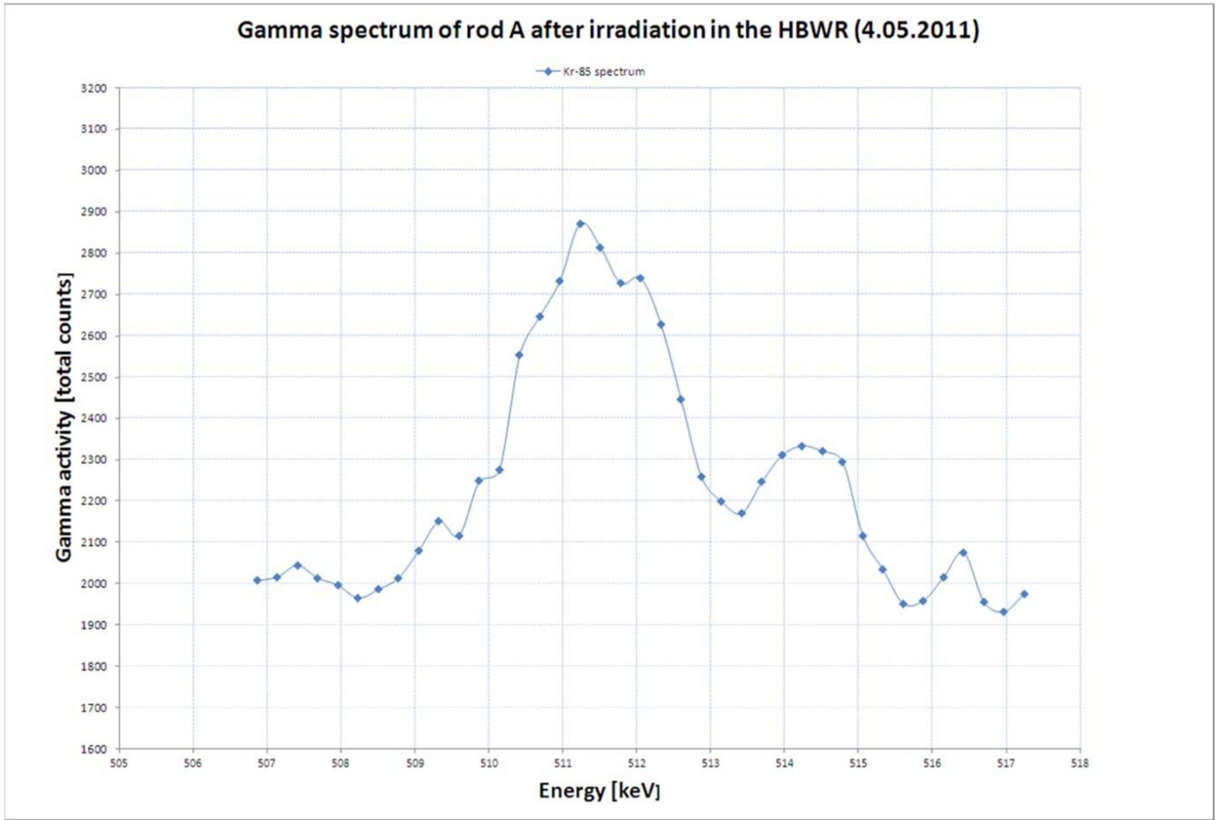
Gamma Scanning Bench Outline



Gamma scanning of fuel stack



Gamma spectrum acquisition at the mid plenum



Measurements of intensity (cps), escape probabilities & production/decay calculations

- UO_2 fuel rod with 16% U-235 enrichment irradiated in 4 cycles up to 58 MWd/kgUO_2 and 5.5 years irradiation time.
- 8.36 mm and 9.5 mm fuel and clad diameter respectively
- 1.307 cm^3 free volume and 15 cm^3 fuel volume
- Collimator 0.3 mm width and 8 mm lead shielding slab in front of the detector
- 25.438 cps & 0.030 cps for 661.66 keV (fuel stack) and for 514 keV (plenum) for rod A
- 0.133 / 0.194 for 514 keV / 661.66 keV escape probability values calculated with microShield software
- 0.782 and 0.914 fractions of Kr-85 and Cs-137 which is not decayed
- FGR (%) = $\frac{R_{Kr85} \times V_{free-volume} \times \gamma_{Cs137} \times D_{Cs137} \times \lambda_{Cs137} \times A_{Cs137} \times B_{Cs137} \times E_{Cs137}}{R_{Cs137} \times V_{fuel} \times \gamma_{Kr85} \times D_{Kr85} \times \lambda_{Kr85} \times A_{Kr85} \times B_{Kr85} \times E_{Kr85}}$ X100%
- $\left[\frac{0.030}{24.9}\right] \times \left[\frac{1.307}{15.0}\right] \times \left[\frac{6.22}{0.27}\right] \times \left[\frac{0.9143}{0.7824}\right] \times \left[\frac{10.78}{30.07}\right] \times \left[\frac{85.2}{0.434}\right] \times \left[\frac{0.133}{0.194}\right] \times \left[\frac{0.852}{1}\right] \times 100\% \cong 11.3\%$

Summary

- FGR (%) obtained for rod A is in very good agreement with puncturing and other destructive examinations performed.
- GammaVision from ORTEC is a good software for calculation of cps at the Kr-85 peak (514 keV) because it can handle overlapping peaks.
- MicroShield software from Grove is an excellent tool for simulation and calculation of escape probabilities of y-rays interacting with matter.
- Method 1 involves only the fuel rod which is to be measured for fission gases.
- Method 2 involves in addition to the rod to be measured a fuel calibration rod. However, it is possible to combine methods 1 and 2 for calculation of absolute efficiencies and other data, e.g. Xe/Kr amount.
- Further experiences must be gained for more extensive analysis of measurement accuracies of the Kr-85 methods.
- Acknowledge to Studsvik and SCK-CEN for basic work performed earlier on the Kr-85 methods for FGR (%) measurements.