

Gamma-ray spectroscopy measurements and simulations for uranium mining

T. Marchais, B. Pérot, C. Carasco, P-G Allinei, P. Chaussonnet, J-L Ma, H. Toubon

Abstract— AREVA Mines and the Nuclear Measurement Laboratory of CEA Cadarache are collaborating to improve the sensitivity and precision of uranium concentration evaluation by means of gamma measurements. This paper reports gamma-ray spectra, recorded with a high-purity coaxial germanium detector, on standard cement blocks with increasing uranium content, and the corresponding MCNP simulations. The detailed MCNP model of the detector and experimental setup has been validated by calculation vs. experiment comparisons. An optimization of the detector MCNP model is presented in this paper, as well as a comparison of different nuclear data libraries to explain missing or exceeding peaks in the simulation. Energy shifts observed between the fluorescence X-rays produced by MCNP and atomic data are also investigated. The qualified numerical model will be used in further studies to develop new gamma spectroscopy approaches aiming at reducing acquisition times, especially for ore samples with low uranium content.

Index Terms— Nuclear measurements, gamma-ray spectroscopy, germanium detector, uranium mining, MCNP6

I. INTRODUCTION

Gamma logging for uranium exploration is currently based on total counting with Geiger Müller gas detectors or NaI(Tl) scintillators. However, the total count rate interpretation in terms of uranium concentration may be impaired in case of roll fronts, when the radioactive equilibrium of the natural ^{238}U radioactive chain is modified by differential leaching of uranium and its daughter elements (Th, Ra, Rn, etc.). Indeed, in the case of secular equilibrium, more than 95% of gamma rays emitted by uranium ores come from ^{214}Pb and ^{214}Bi isotopes, which are in the back-end of ^{238}U chain. Consequently, these latter might still produce an intense gamma signal even when uranium is not present in the ore, or in smaller quantity than in the secular equilibrium. Therefore, additional gamma measurements of core samples are performed in surface with high energy resolution, high-purity germanium (HP Ge) detectors. These last allow the detection of imbalances using the ratio of different gamma lines of the ^{238}U decay chain, such as the well-know 1001 keV of $^{234\text{m}}\text{Pa}$, one of the first daughters in the beginning of the chain. However due to the low intensity of this gamma ray (0.84%),

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its detection in ore samples of a few hundred grams with a low uranium concentration may require measurement times of several hours. Therefore, we plan to use more intense peaks at lower energy, such as self- fluorescence X rays of uranium, to reduce acquisition times. Fluorescence is induced by the main gamma radiations emitted in the ore by ^{214}Pb (242, 295, 352 keV), ^{214}Bi (609 keV) and ^{226}Ra - ^{235}U (186 keV). After Compton scattering, these gamma rays produce a high continuum of lower energy photons in the K-edge region of uranium (115.6 keV), resulting in a large photoelectric absorption rate and, subsequently, in the emission of uranium fluorescence X-rays, like the most intense 98.4 keV line. In order to study the potential of these new approaches and to establish a numerical simulation model, a measurement campaign was carried out in the radiometric calibration station of AREVA Mines in Bessines, France [1], with a coaxial HPGe detector. This station holds seven cement blocks with increasing uranium contents, up to 1% weight fraction (i.e. 10,000 ppm). The optimization of the MCNP model of the HPGe detector will be presented, as well as a comparison of different nuclear data libraries to explain missing or exceeding peaks in the simulation. Energy biases between the fluorescence X-rays produced by MCNP and atomic data will also be investigated.

II. THE RADIOMETRIC CALIBRATION STATION

AREVA Mines radiometric calibration station in Bessines aims to measure the counting rate due to gamma radiation emitted by seven independent cubic standard blocks with 70 cm edges (Fig.1). The uranium content of each block has been determined by chemical analysis (Table I). These blocks allow the calibration of radiometric probes.

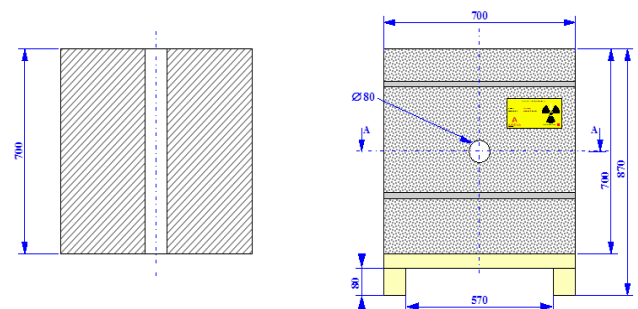


Fig. 1. Drawing of a standard block [1]

TABLE I. CHARACTERISTICS OF THE SEVEN BLOCKS

Block #	Uranium mass fraction from chemical analyses (ppm)	Cement density (g.cm ⁻³)
B1	0	1.89
B2	500	1.92
B3	1000	1.89
B4	1900	1.91
B5	2900	1.92
B6	4800	1.93
B7	9700	1.92

The Nuclear Measurement Laboratory of CEA Cadarache has carried out a series of gamma-ray spectroscopy measurements on these standard blocks (Fig. 2). Results have confirmed the uranium content of each block, as shown further in Table II. The gamma spectra acquired during this campaign (Fig. 3) will also be used to validate the MCNP numerical model and to identify new information (X or γ peaks, Compton continuum, K-edge, etc.) that could be used to improve the assessment of uranium concentration. This campaign has been performed with a type N, coaxial HPGe detector, model GR1020, CANBERRA.



Fig. 2. Coaxial detector inside a standard cement blocks at Bessines.

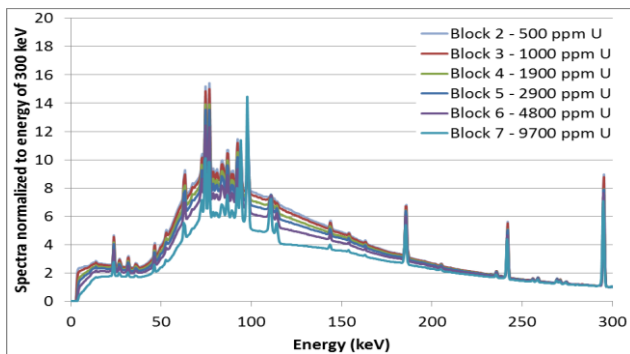


Fig. 3. Gamma-ray spectra of the seven calibration blocks of AREVA radiometric station in Bessines, France.

III. SIMULATION OF THE COAXIAL DETECTOR

The coaxial detector was modeled with the MCNP Monte Carlo computer code [2]. The geometric model of the detector is based on the manufacturer scheme, on X-ray radiography, and on measurements performed with a multi-energy beam coming from a highly collimated ¹⁵²Eu gamma source (see Fig. 4), which allowed to precisely estimate the position and active area of the crystal. The calculated and experimental

detector efficiencies are reported in Fig. 5. The measured efficiency was built with calibration point sources (²⁴¹Am 59.5 keV line, ¹⁰⁹Cd 88.0 keV peak, and ¹⁵²Eu gamma-rays from 121.8 keV to 1408 keV) at a distance of 50 cm from the detector head. Note that to obtain the good agreement observed in Fig. 5, a dead layer has been implemented in the model of the germanium crystal, in the back, external edge of the germanium cylinder, to reflect a deficit of charge collection in this region. A copper electrode connecting the cooling system and the germanium crystal is also described, as well as the external aluminum cover.

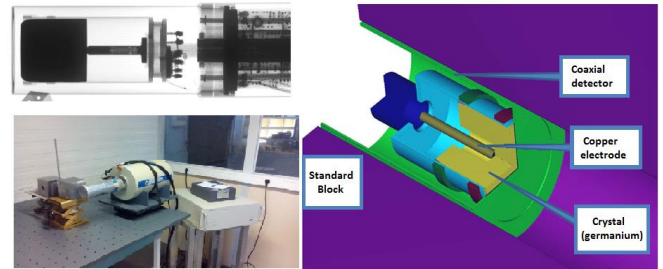


Fig. 4. X-ray radiography and the modeling of the coaxial detector.

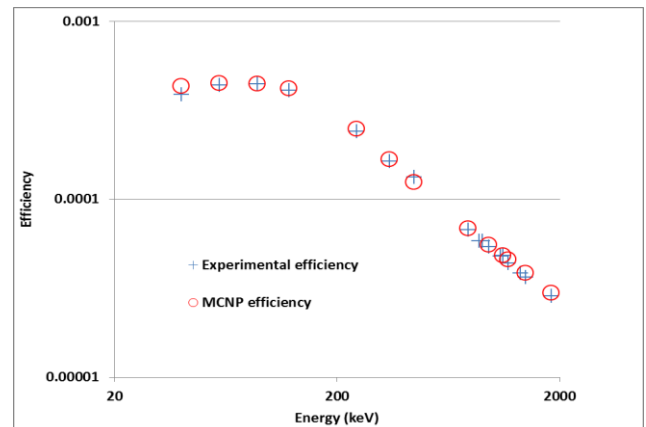


Fig. 5. Experimental and MCNP efficiency of the coaxial HPGe measured with point sources at a distance of 50 cm from the detector entrance face.

IV. VERIFICATION OF THE URANIUM CONTENT IN THE STANDARD CONCRETE BLOCKS

The global detection efficiency, including self-absorption in the standard cement blocks, has been calculated with MCNP with a first objective to determine the uranium mass concentration of each block, using the ^{234m}Pa gamma ray at 1001 keV. Equation (1) allows converting the net area of this peak into uranium mass concentration (ppm).

$$Cm_U = \frac{(1 + \eta) \times 10^6 \times M_{mol}({}^{238}U) \times T_{1/2}({}^{238}U)}{N_A \times \ln(2)} \times \frac{S_n(1001 \text{ keV})}{Tc \times M_{Block} \times I_\gamma(1001 \text{ keV}) \times Eff(1001 \text{ keV})} \quad (1)$$

With:

Cm_U : Uranium mass concentration of each block (ppm)

$S_n(1001 \text{ keV})$: Net area of the peak at 1001 keV measured on the gamma spectrum (number of counts, dimensionless number)

T_C : Acquisition “live time” (i.e. real time corrected for dead time) of the gamma spectrum (s)

$I_\gamma(1001\text{ keV})$: Intensity of emission of the peak at 1001 keV of ^{234m}Pa (0.84 %) in the ^{238}U filiation

$Eff(1001\text{ keV})$: Detection efficiency calculated with MCNP for each block (number of count in the full-energy peak at 1001 keV per photon of 1001 keV emitted in the block)

$T_{1/2}(^{238}\text{U})$: Radioactive period of ^{238}U (s)

N_A : Avogadro constant ($6,023 \cdot 10^{23}$ atoms per mole, dimensionless number)

$M_{mol}(^{238}\text{U})$: Molar mass of ^{238}U (g)

η : Mass fraction of ^{235}U in natural uranium (0.72 %)

The mass concentration obtained by gamma-ray spectroscopy using (1) is in good agreement with chemical analysis, as shown in Table II. The reported uncertainty includes the detector calibration and modeling uncertainty (2.6 %), the statistical uncertainty on the net area of the peak at 1001 keV (between 0.9 % and 22 %, depending on uranium concentration), the uncertainty on the 1001 keV gamma-ray intensity (1.32 %), the statistical uncertainty of MCNP calculations (less than 1 %), and the uncertainty of the MCNP modeling of the concrete blocks and measurement setup. Concerning this last, the main source of uncertainty concerns concrete density, which varies from 1.89 to 1.93 g.cm⁻³ depending on the measured block. The dispersion of the efficiency values calculated on the blocks with MCNP is equal to 0.97 %. This value has been retained to be the uncertainty of the modeling of the measurement scene at 1 σ . The global uncertainty is the quadratic sum of these uncertainties.

TABLE II. URANIUM MASS CONCENTRATIONS (PPM) OF BESSINES CALIBRATION BLOCKS MEASURED BY GAMMA-RAY SPECTROSCOPY AND CHEMICAL ANALYSIS

Block	Chemical analysis	Gamma-ray spectroscopy (and 1 σ uncertainty)	Relative difference
B1	0	2.1 (\pm 22 %)	N/A
B2	500	459 (\pm 3.7 %)	- 8.2 %
B3	1000	958 (\pm 1.9 %)	- 4.2 %
B4	1900	1960 (\pm 2.3 %)	3.4 %
B5	2900	2800 (\pm 3.3 %)	- 3.5 %
B6	4800	4550 (\pm 1.8 %)	- 5.2 %
B7	9700	9530 (\pm 2.0 %)	- 1.8 %

V. SIMULATION OF THE STANDARD CONCRETE BLOCKS GAMMA SPECTRA

The simulation of the concrete blocks is based on a cement chemical composition given by AREVA Mines, including uranium concentrations (and densities) given in Table I. An isotropic gamma source uniformly distributed in the cement block is implemented in MCNP. The natural ^{238}U and ^{235}U radioactive chains are supposed in secular equilibrium. The source term retains only 14 predominant gamma emitters observed in the experimental spectra: ^{238}U , ^{234}Th , ^{234m}Pa ,

^{230}Th , ^{226}Ra , ^{214}Pb , ^{214}Bi , ^{210}Pb , ^{235}U , ^{231}Th , ^{227}Th , ^{223}Ra , ^{219}Rn , and ^{211}Pb . The intensity and energy of each gamma ray are taken from the JEFF 3.1.1 library, and only the lines with energy larger than 40 keV are modeled.

Fig. 6 shows a good general agreement between simulated and experimental spectra, only small differences being observed for minor peaks. For instance, the 89.5 keV peak was initially absent from the simulated spectrum due to a missing emission line of ^{214}Pb at 89.8 keV in the JEFF 3.1.1 library (Bi $K_{\beta 2}$ X-ray, intensity of 0.67% in ENSDF [3]), combined with an underestimation emission of ^{214}Bi at 89.2 keV (Po $K_{\beta 3}$ X-ray, 0.085% in JEFF 3.1.1 instead of 0.116% in ENSDF [3]). After implementing ENSDF data [3], the calculated 89.5 keV peak was found in satisfactory agreement with experiment (Fig. 7).

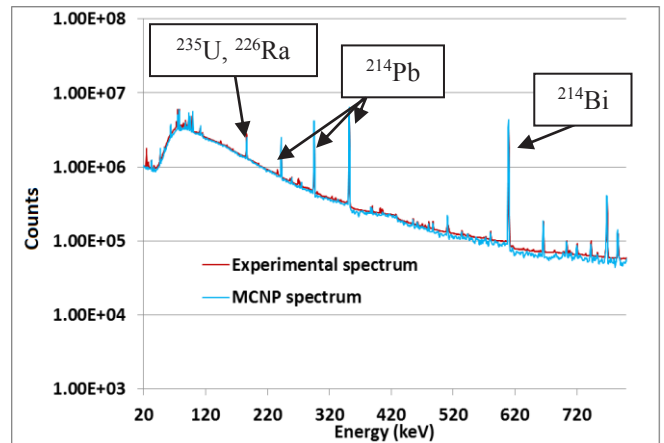


Fig. 6. Experimental and MCNP spectra of block B5 using ENSDF database.

We can also note in Fig. 7 that a peak is missing in the simulation at 110.4 keV, in the broad bump including both 110.4 and 111.3 keV uranium fluorescence X-rays ($K_{\beta 3}$ and $K_{\beta 1}$, respectively [4]). Indeed, MCNP manual [5] indicates that for elements with atomic number $Z > 37$, the X-ray emission $K_{\beta 3}$ is not simulated, and $K_{\beta 1}$ X-ray includes all transition intensities from layers L to K, which explains the absence of the 110.4 keV peak ($K_{\beta 3}$) and a higher intensity at 111.3 keV ($K_{\beta 1}$).

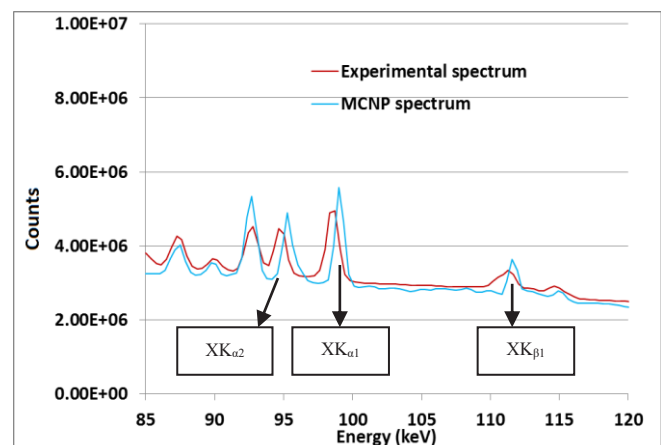


Fig. 7. Experimental and MCNP spectra of block B5 at low energy.

Fig. 7 also shows a significant energy shift between calculation and experiment concerning uranium fluorescence

X-rays. Further investigation of the MCNP fluorescence peaks energy shift has been performed for 53 materials having atomic numbers between 40 (zirconium) and 92 (uranium) irradiated by a 1 MeV photon beam. The energy shift between MCNP and the expected fluorescence X-ray energy shown in Fig. 8 as a function of Z reproduces the observation previously made in [6].

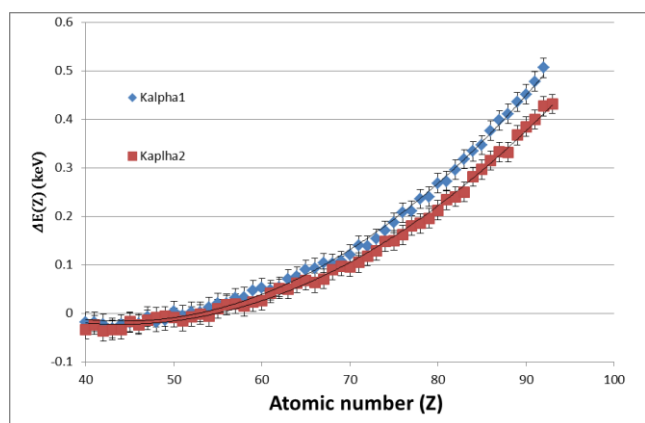


Fig. 8. Energy shift between the energy of fluorescence X-rays calculated with MCNP and the theoretical values taken from [3].

In further studies, this energy shift will be corrected when processing MCNP output data to allow a better agreement with the experimental spectrum.

VI. CONCLUSION AND PROSPECTS

Seven calibration blocks with known uranium concentrations have been measured by high resolution gamma-ray spectroscopy at AREVA Mines calibration station, in Bessines, France, and simulated with MCNP computer code. First, a detailed model of the coaxial HPGe detector has been carried out using calibration acquisitions with standard point sources of ^{109}Cd , ^{152}Eu , and ^{241}Am . Then, the uranium mass concentrations of the blocks (in ppm), originally determined by chemical analysis, has been confirmed by gamma-ray spectroscopy with the 1001 keV gamma ray of $^{234\text{m}}\text{Pa}$, in the ^{238}U radioactive chain. Finally, we obtained a very good agreement between the overall gamma spectra of the blocks calculated with MCNP and the ones measured in Bessines. Only minor discrepancies concerning small peaks have been observed and corrected, which were due to unprecise or missing nuclear data, as well as small bias in the energy of fluorescence X-rays produced by MCNP, which will be corrected in future studies.

We will now use the validated numerical model to study new gamma-ray spectroscopy methods, in view to characterize uranium faster than with the reference 1001 keV line. Indeed, its intensity (0.84 %) leads to hours of acquisition time for ore samples of a few hundred grams with uranium concentration lower than 1 % mass fraction. Among the information that is not yet used, uranium self-fluorescence X-rays in the 100 keV region (K_{α} and K_{β} transitions) can be detected in a much shorter time. They are induced by gamma radiations of the ore itself and could be used to characterize uranium up to a depth of several centimeters, hence being appropriate for small ore samples.

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