Experimental validation of a $CeBr_3$ gamma-ray logging probe MCNP model

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Abstract— Orano Mining has relied on Nuclear Measurement Laboratories for several years to estimate calibration factors for borehole radiometric probes. The total gamma count rate recorded with a NaI(Tl) scintillation detector (NGRS probe) is converted into equivalent uranium grade using a calibration coefficient in $s^{-1}.ppm_U^{-1}$ units, estimated thanks to different calibration blocks ranging from 0 to 10,000 ppm of uranium, at Orano CIME calibration facility in Bessines, France. Recently, Orano Mining embarked on an update of its measurement means by associating gamma spectrometry directly in the wellbore. Therefore, Orano Mining has recently tested a $CeBr_3$ probe designed and developed by ALT (Advanced Logic Technology) to qualify a patented borehole spectroscopic method based on energy bands recently developed by CEA and Orano. In this context, we have implemented a fine Monte Carlo modelling of this new probe in order to correct the calibration coefficients as a function of different parameters such as the drilling diameter, the presence of a casing, the density of the mineralization, the distance of the probe with respect to the well or casing walls, etc. In the present work, we have simulated the $CeBr_3$ probe in the concrete calibration blocks of Orano CIME. The MCNP model of the probe has been validated through a comparison with experimental data. The calibration coefficient determined by simulation is 11.6 $s^{-1}.ppm_U^{-1}$ ± 10 % in total gamma counting, which is in good agreement with the one measured in Bessines, i.e. 10.6 $s^{-1}.ppm_U^{-1}$. The calculation vs. experiment agreement is also satisfactory (i.e. within 10 %) for spectroscopic energy bands used to characterize the uranium grade in case of disequilibrium in the uranium chain, when total count rate does not apply. This method recently developed for samples and boreholes will be tested in future work with this new $CeBr_3$ probe.

Index Terms— $CeBr_3$(Ce) probe for uranium borehole logging, MCNP simulation, experimental calibration

I. Introduction

The development of the $CeBr_3$ probe was entrusted by Orano Mining to the company Advanced Logic Technology (ALT [1]). The $CeBr_3$ crystal has dimensions of 2.0 cm in diameter and 9.6 cm in length. The $CeBr_3$ probe was simulated with the MCNP6 computer code [2] using data provided by ALT (see Fig.1).

The main emissions of uranium ore come from ^{238}U, ^{235}U and ^{232}Th decay chains shown in Fig. 2, and from ^{40}K. The majority of the gamma signal comes from ^{214}Bi and ^{214}Pb located at the end of the ^{238}U decay chain.

It should be noted that the decay chains may be subject to possible radioactive imbalances resulting from geochemical phenomena (such as differential leaching of uranium and radium in roll-fronts) or from physicochemical treatments on the ore (radon evaporation during crushing or drilling). Indeed, uranium is a mobile element in the ore and its mobility is influenced by the chemical conditions of the environment. The main imbalance considered during mining prospection is between uranium and radium, denoted U/Ra (activity ratio). As the majority of gamma emissions come from ^{214}Pb and ^{214}Bi, the uranium content estimated in total count rate, such as the NGRS probe [3] (Natural Gamma Ray Probe), might be erroneous without the knowledge of the imbalance between the top and bottom activities of the ^{238}U decay chain. Therefore, we developed new gamma spectroscopy approaches for a $LaBr_3$(Ce) probe prototype [4], which will be now tested with the new industrial $CeBr_3$ probe.

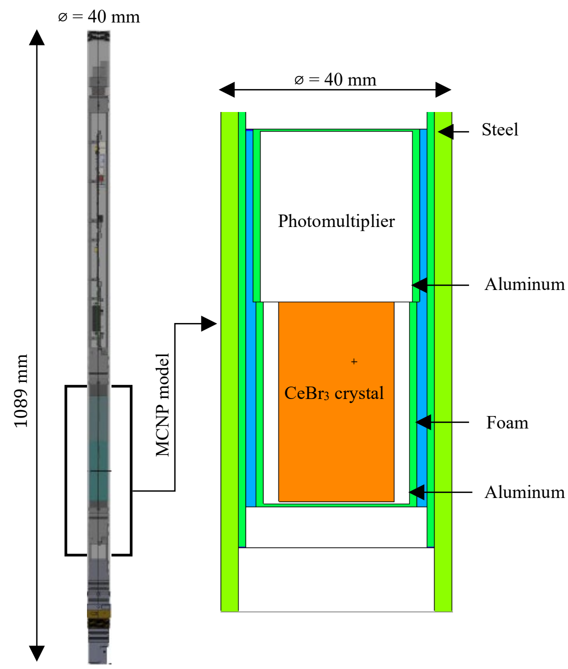

Fig. 1. QL40-SGR-2G probe from Advanced Logic Technology (ALT) and its associated MCNP model.

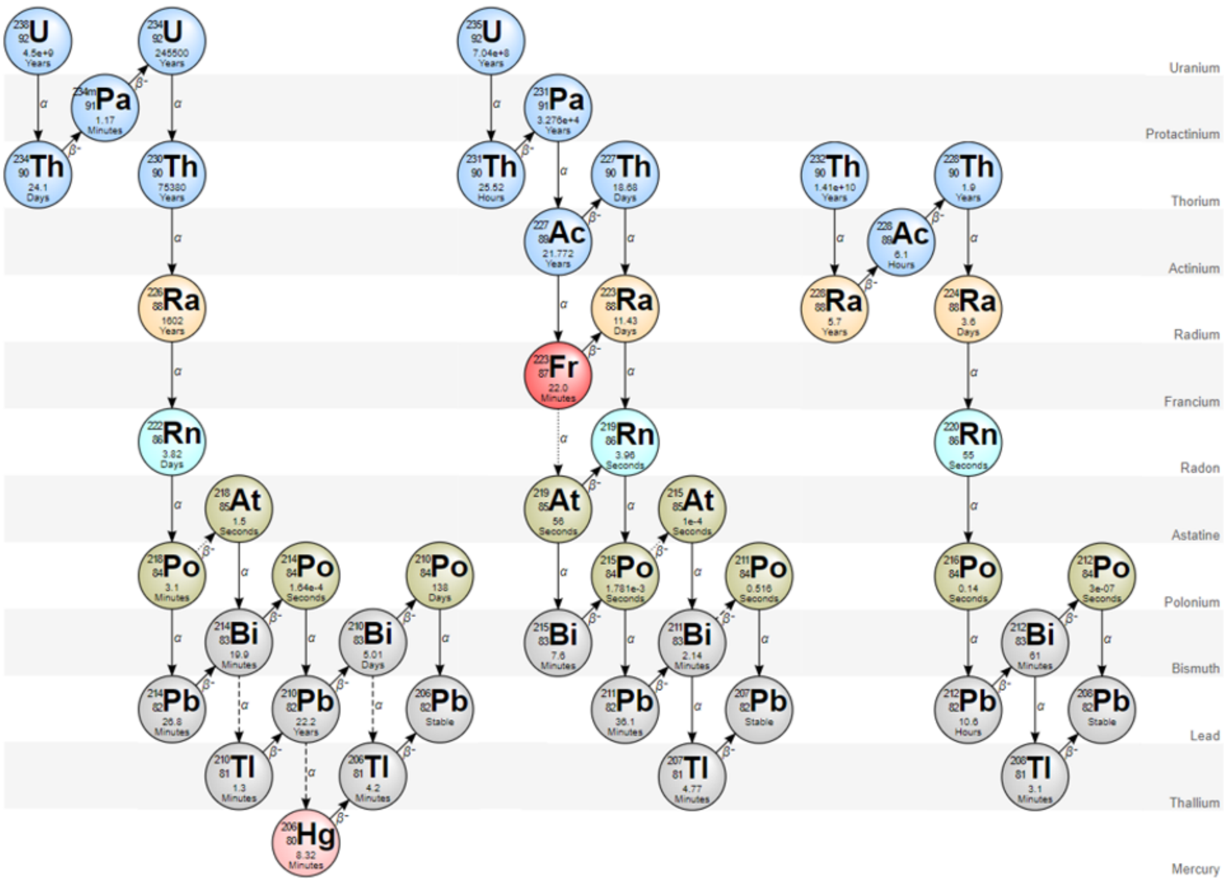

Fig. 2. Decay chains of ^{238}U, ^{235}U and ^{232}Th and associated radioactive periods [5].

II. CeBr$_3$ probe simulation in calibration blocks

Foremost, the experimental calibration of this probe in the Sosvurt block (located in Kazakhstan) and in the blocks of Orano CIME calibration station (in Bessines, France) made it possible to acquire gamma spectra with uranium contents ranging from 500 to 9700 ppm$_U$.

The CeBr$_3$ probe was initially simulated in the Sosvurt block with a content of 1000 ppm$_U$, see Figure 3. The thorium and potassium contents of this block were estimated by Orano Mining at respectively 20 ppm and 3.7 % (mass fractions). The spectrum obtained by MCNP simulation is compared to the spectrum obtained experimentally, see figure 4.

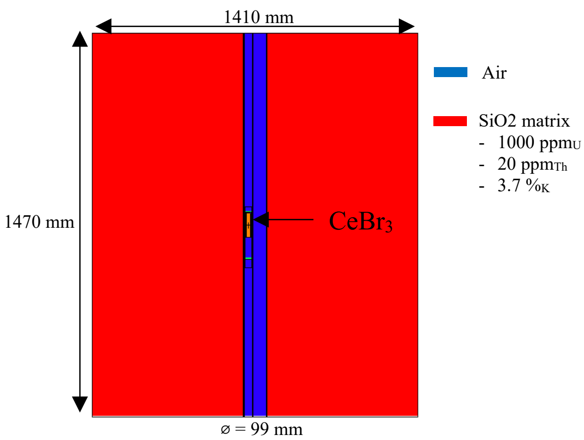

Fig. 3. MCNP modelling of CeBr$_3$ probe in Sosvurt block.

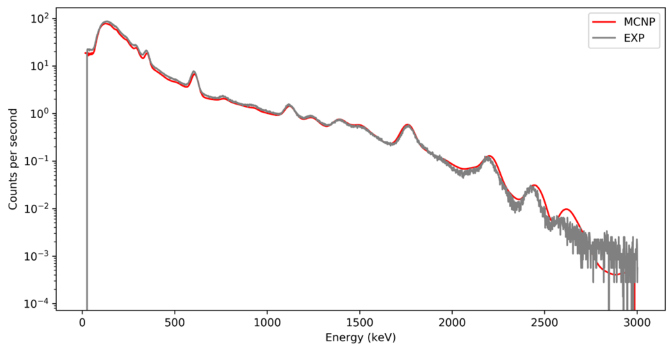

Fig. 4. Simulated and measured spectra of CeBr$_3$ probe in Sosvurt block.

A good agreement is observed, with only a 5.0 % overestimation of the experimental total count rate (9770 s^{-1}) by numerical simulation (10240 s^{-1}) for an energy larger than the electronics low-energy threshold of 80 keV.

The experiment-calculation comparison was then performed with gamma spectra measured in the calibration blocks of Bessines, with a uranium content ranging from 500 to 9700 ppm$_U$. The thorium and potassium contents of these blocks were estimated by Orano Mining at 15 ppm$_{Th}$ and 3.6 % (mass fractions), respectively. Figure 5 shows the MCNP model of the CeBr$_3$ probe in a Bessines block and Figure 6 shows the spectra obtained by numerical simulation and experiment for the 2900 ppm$_U$ block.

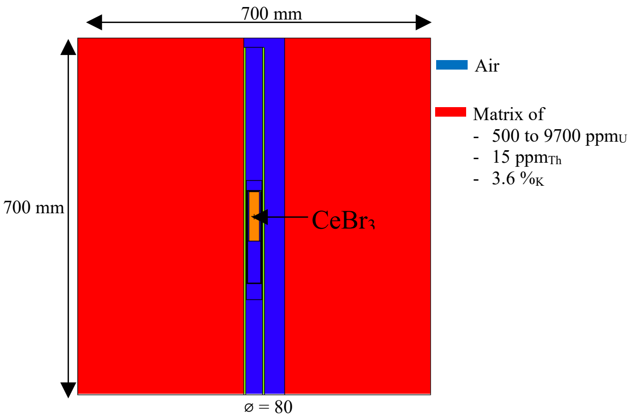

Fig. 5. MCNP modelling of $CeBr_3$ probe in Bessines block.

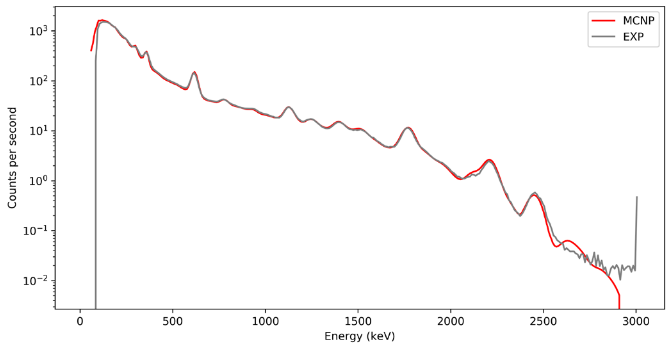

Fig. 6. Simulated and measured spectra of CeBr3 probe inside Bessines block with a 2900 ppm_U grade.

Table I reports, for each of the six Bessines blocks, the difference between experiment and simulation on the total counts recorded above the low-energy threshold of 80 keV. An average relative difference of 7 % is observed between numerical simulation and experiment. This value is similar to that of the Sosvurt reference block (+ 5 %).

TABLE I
TOTAL COUNTING RATES, C_{TOT}, OBSERVED EXPERIMENTALLY AND BY NUMERICAL SIMULATION FOR ENERGY GREATER THAN 80 KEV

Block	U grade (ppm)	C_{TOT}^{EXP} (s^{-1})	C_{TOT}^{MCNP} (s^{-1})	$\frac{MCNP - EXP}{EXP}$
B2	500	5356	5878	+ 9.7 %
B3	1000	10620	11515	+ 8.4 %
B4	1900	21155	21452	+ 1.4 %
B5	2900	30169	32188	+ 6.7 %
B6	4800	48772	51789	+ 6.2 %
B7	9700	89872	98191	+ 9.3 %

Fig. 7, shows the total counting rate C^{TOT} as function of the uranium grade of Bessines blocks.

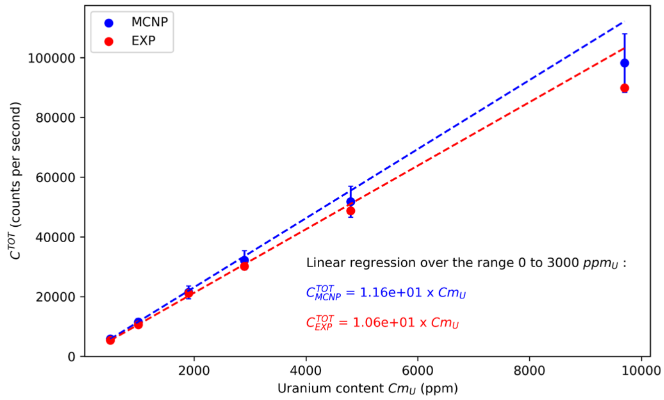

Fig. 7. Total counting rate C_{TOT} observed experimentally and by numerical simulation for energy greater than 80 keV as function of uranium grade (ppm). The linear regression fits only includes the data from 0 to 3000 ppm_U to avoid the saturation effects mentioned in the text.

The uncertainty plotted for MCNP simulation (10 %) includes those due to detector modelling, statistical uncertainty of MCNP calculations and knowledge of the standard blocks [6]. A beginning of saturation is observed both in MCNP simulation, due to gamma self-absorption in uranium [3], and in the experiment due to self-absorption but also electronics losses in the highest count rates.
In order to use the K_{APC} indicator of uranium content developed in previous study [4] [5] and described in section III, we also compare in Table II the experimental and calculated count rates in two energy areas (120-170 keV and 960-1040 keV), which evidences a slight distortion of the spectra: MCNP globally overestimates the low-energy signal but underestimates the high-energy count rate for most of the blocks.

TABLE II
COUNT RATES OBSERVED EXPERIMENTALLY AND BY NUMERICAL SIMULATIONS BETWEEN 120-170 KEV AND 960-1040 KEV

	Count rates between 120-170 keV			Count rates between 960-1040 keV		
Block	EXP (s^{-1})	MCNP (s^{-1})	$\frac{MCNP - EXP}{EXP}$	EXP (s^{-1})	MCNP (s^{-1})	$\frac{MCNP - EXP}{EXP}$
B2	1361	1466	+ 7.7 %	32	34	+5.1 %
B3	2690	2855	+ 6.1 %	66	67	+0.8 %
B4	5256	5242	- 0.3 %	136	125	- 8.0 %
B5	7326	7756	+ 5.9 %	198	190	- 4.2 %
B6	11438	12144	+ 6.2 %	334	313	- 6.2 %
B7	19202	21632	+ 12.7 %	693	631	- 9.0 %

III. Indicators of the uranium grade

The traditional approach based on the total count rate uses a CC_{TOT} calibration coefficient, which is the slope of the curves of Fig. 7. This coefficient links the total count rate above the low-energy threshold of 80 keV, C_{TOT}, and the uranium content:

$$Cm_U = \frac{C_{TOT}}{CC_{TOT}} \quad (1)$$

Their values are CC_{TOT}^{EXP} = 10.6 c.s^{-1}.ppm$_U^{-1}$ and CC_{TOT}^{MCNP} = 11.6 c.s^{-1}.ppm$_U^{-1}$ (see Fig. 7), with an uncertainty estimated to 10 %. These uncertainties include those due to detector modelling, statistical uncertainty of MCNP calculations, knowledge of the standard blocks [6] (uranium grade, density, concrete composition), and also the precision on the energy of the electronics threshold.

On the other hand, the new spectroscopic approach described in [4] and [5] uses another indicator K_{APC} defined as the ratio between the counting rates in the 120 – 170 keV and 960 – 1040 keV areas. K_{APC} is related to the uranium content through the following formula:

$$Cm_U = \frac{\beta - K_{APC}}{\alpha} \quad (2)$$

with:

- $K_{APC} = \frac{C_{[120-170]}}{C_{[960-1040]}}$, $C_{[120-170]}$ and $C_{[960-1040]}$ being the number of counts in the abovementioned energy bands,
- α and β the parameters of the linear relationship between K_{APC} indicator and Cm_U uranium content, established with the data of Table II, see Fig. 7.

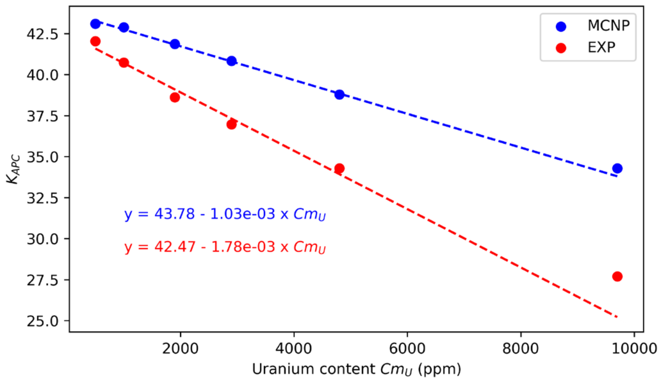

Fig. 8. K_{APC} indicator as function of uranium content in Bessines blocks.

As in Fig. 7 for the total count rate indicator, the linear fits reported on Fig. 8 only use the points below 5000 ppm_U. Indeed, regardless a possible spectrum distortion between the low and high energy areas at high count rate, the attenuation of the Compton continuum in the low-energy band, which is due to gamma self-absorption in uranium, starts saturating above 5000 ppm_U. The β coefficients are quite similar in MCNP and experiment, but the slopes are quite different, probably due to the gamma spectrum distortion that can be observed in the data of Table II. Therefore, the new $CeBr_3$ probe will be sent to the Nuclear Measurement Laboratory of CEA to investigate the effect of increasing count rates on the shape of the gamma spectrum and on count losses.

IV. Conclusion

The new $CeBr_3$ probe developed by ALT was simulated by CEA with MCNP6 computer code and its model was qualified using experimental data provided by Orano Mining. The probe modelling in the Sosvurt block and in the Bessines calibration blocks made it possible to observe the influence of the uranium content (from 500 to 9700 ppm_U) on the gamma spectra and on two uranium indicators: on the one hand the total count rate above the 80 keV low-energy threshold, and on the other hand the K_{APC} ratio between two energy bands of the spectrum (120-170 keV and 960-1040 keV, respectively). We observe a good agreement between simulation and experiment on the total count rate, but the expected self-attenuation of the Compton continuum in the 120-170 keV band seems to be underestimated in numerical simulations. In fact, we rather suspect count losses at low energy when the total count rate increases along with uranium content. This hypothesis will be investigated in further studies when the $CeBr_3$ probe will be sent by Orano Mining to the Nuclear Measurement Laboratory of CEA for a detailed characterization, both to refine the modelling of the crystal and its probe environment, but also to investigate count-rate related spectrum distortions and losses.

References

[1] “ALT site web,” *ALT*. https://www.alt.lu/ (accessed Feb. 22, 2023).

[2] Los Alamos National Laboratory, “MCNP6, User's manual – Version 1.0 - LA-CP-13-00634, Rev. 0 – May 2013 – Denise B. Pelowitz, editor.” 2013.

[3] C. Carasco, B. Perot, J.-L. Ma, H. Toubon, and A. Dubille-Auchere, “Improving Gross Count Gamma-Ray Logging in Uranium Mining With the NGRS Probe,” *IEEE Trans. Nucl. Sci.*, vol. 65, no. 3, Art. no. 3, Mar. 2018, doi: 10.1109/TNS.2018.2800909.

[4] Bertrand Pérot, Thomas Marchais, Pierre-Guy Allinei, Hervé Toubon, Youcef Bensedik, Romain Goupillou, Alexandre Berland, "Development of Gamma Spectroscopic Tools for Uranium Ore Samples and Borehole Exploration", 2022 IEEE Nuclear Science Symposium, 5 – 12 November 2022, Milano, Italy.

[5] T. Marchais, B. Perot, C. Carasco, H. Toubon, and Y. Bensedik, “Method for assessing the uranium content using gamma spectrometry in a wellbore and associated device,” WO2021122772 (A1), Jun. 24, 2021 Accessed: Oct. 25, 2021. [Online]. Available: https://worldwide.espacenet.com/publicationDetails/biblio?FT=D&date=20210624&DB=EPODOC&locale=fr_EP&CC=WO&NR=2021122772A1&KC=A1&ND=4

[6] T. Marchais *et al.*, “Detailed MCNP Simulations of Gamma-Ray Spectroscopy Measurements With Calibration Blocks for Uranium Mining Applications,” *IEEE Trans. Nucl. Sci.*, vol. 65, no. 9, pp. 2533–2538, Sep. 2018, doi: 10.1109/TNS.2018.2797312.