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# Development of gamma spectroscopic tools for uranium ore samples and borehole exploration

Bertrand Pérot, Thomas Marchais, Pierre-Guy Allineï  
Hervé Toubon, Youcef Bensedik, Romain Goupillou, Alexandre Berland

**Abstract**— Measuring uranium ore samples with high resolution gamma spectrometers requires long acquisition times and expensive high-purity germanium (HPGe) detectors that are difficult to operate in the field (crystal cooling at very low temperature). In this work, we present a spectrometer based on a  $1.5'' \times 1.5''$  LaBr<sub>3</sub>(Ce) scintillator able to measure, in a few minutes, the uranium concentration and the possible disequilibrium of the uranium chain. The method is based on the total counts in two energy bands of the gamma spectrum, of which one at low energy that is sensitive to the top of the  $^{238}\text{U}$  chain (including  $^{234}\text{Th}$  gamma rays near 92 keV and the 98.4 keV X-ray due to uranium self-induced fluorescence). The second band, around the 609 keV peak of  $^{214}\text{Bi}$  (bottom of the chain), is insensitive to uranium and is used for normalization. We will also present the development of a borehole logging probe based on a LaBr<sub>3</sub>(Ce) detector and two energy-band approaches, first with a narrower low-energy band focused on the 98.4 keV X-ray and the same normalization with the 609 keV peak. Their ratio is almost insensitive to uranium grade, owing to competitive effects of self-fluorescence and self-absorption, but it gives the radioactive disequilibrium through a linear correlation. In the second approach, the low-energy band is sensitive to the attenuation of the Compton continuum due to uranium, divided by the counts in an energy band encompassing the 1001 keV gamma ray of  $^{234\text{m}}\text{Pa}$ . The ratio is not sensitive to the disequilibrium, but is linear with uranium concentration. Finally, it is also possible to measure the net area of the 1001 keV gamma ray with the LaBr<sub>3</sub>(Ce) probe, at high uranium grade. Among these three approaches, the most efficient one is chosen depending on uranium grade and radioactive disequilibrium. The 1001 keV net area is also used at low grade with a longer acquisition time to self-calibrate the probe in real boreholes.

**Index Terms**— LaBr<sub>3</sub>(Ce) spectrometer for uranium ore samples, LaBr<sub>3</sub>(Ce) probe for uranium borehole logging

## I. INTRODUCTION

**O**RANO Mining and the Nuclear Measurement Laboratory of CEA IRESNE Institute are developing a new gamma spectrometer for uranium ore samples and a logging probe for borehole exploration based on LaBr<sub>3</sub>(Ce) scintillators. High resolution HPGe spectrometers [1] are widely used in the laboratory but low-resolution NaI(Tl) scintillators are preferred in the field to estimate the uranium grade and the potential radioactive disequilibrium between uranium, radium and radon (see Fig. 1) in ore samples [2]. The disequilibrium is caused by the difference in mobility of uranium and radium in roll-fronts deposits [3]. We consider here that radium and radon are in equilibrium as the beakers containing uranium ores have been hermetically closed to prevent radon leakage, more than one

This work has been performed in the frame of an R&D collaboration between CEA and Orano Mining.  
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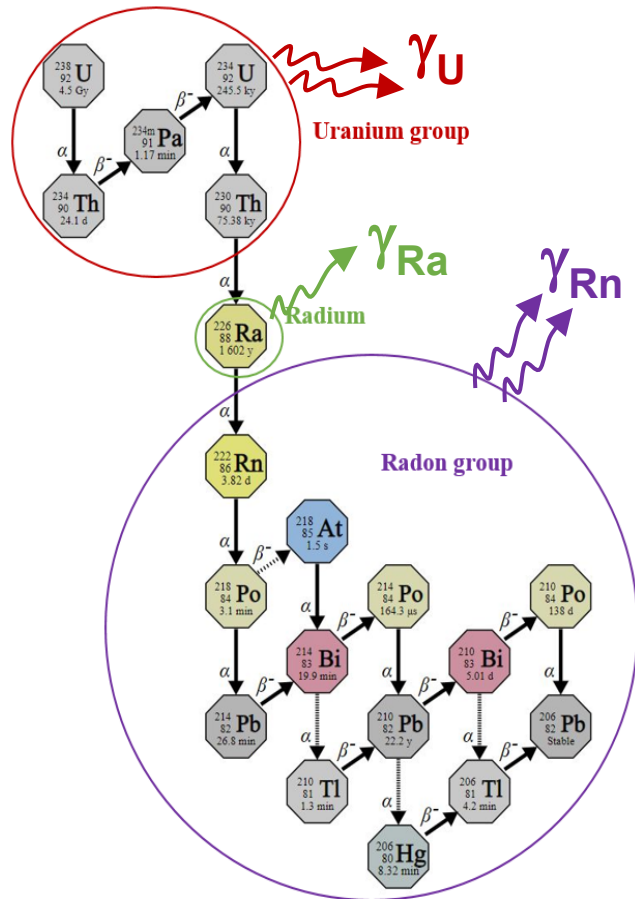


Fig. 1.  $^{238}\text{U}$  decay chain (Wikipedia) in which the circles show the groups of interest in terms of radioactive disequilibrium (see text for details).

month before measurements to allow the equilibrium recovery between  $^{222}\text{Rn}$  (radioactive half-life of 3.8 days) and its daughters. Radon effects are also observed in boreholes just after drilling, with equilibrium recovery after about one month. In this work, we will refer equivalently to U/Ra or U/Rn disequilibrium. This paper reports MCNP6.1 [4] calculations to design and study the uncertainties of these LaBr tools for uranium exploration and exploitation, and their first tests to validate the expected performances.

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## II. LABR SPECTROMETER FOR URANIUM ORE SAMPLES

Monte Carlo simulations with the MCNP6.1 computer code [4] have been performed to optimize the design of the LaBr spectrometer and perform parametric studies on different effects, such as ore density, mineralogy, filling level in the beaker, and gamma self-absorption in uranium. The MCNP model and industrial design of the spectrometer is shown in Fig. 1. It includes a 1.5"×1.5" LaBr<sub>3</sub>(Ce) scintillator from Saint-Gobain Crystals, coupled to CAEN "i-Spector" (Intelligent Silicon Photomultiplier Tube with Digital MCA), a 2.5 cm thick lead shield against external background, and the crushed ore beaker.

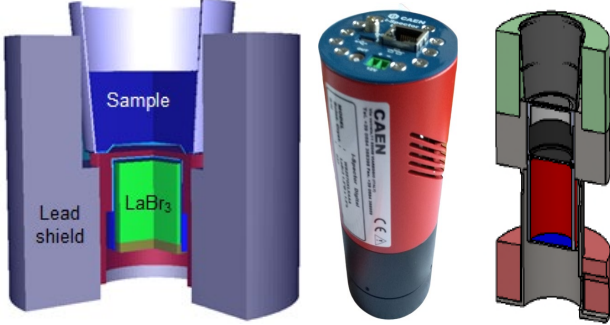


Fig. 2. LaBr spectrometer for uranium ore samples (from left to right the MCNP model, the "i-Spector" module, and the industrial design).

Figure 3 shows the relative contributions of the two main groups of uranium and radon (cf. Fig. 1), with the two energy bands from 87 to 110 keV (gamma rays of <sup>234</sup>Th at 92.4 and 92.8 keV, and K<sub>α1</sub> fluorescence X-ray of uranium at 98.4 keV), and from 560 to 660 keV (609 keV peak of <sup>214</sup>Bi) used to estimate the radioactive disequilibrium and the uranium grade.

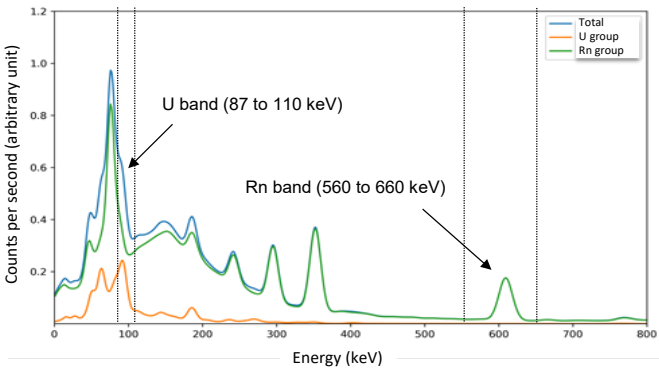


Fig. 3. Simulated contributions of the uranium and radium-radon groups for a reference ore sample with a SiO<sub>2</sub> mineralogy, a 1000 ppm grade of uranium (here with the chain in equilibrium), a filling height of 4 cm, a density of 1.2 g.cm<sup>-3</sup> and a mass of 157 g.

Following the method detailed in [2] and [4], the count ratio of the two energy bands linearly depends on the U/Ra disequilibrium (activity ratio):

$$\frac{C_{U \text{ band}}}{C_{Rn \text{ band}}} = \alpha \frac{A_U}{A_{Rn}} + \beta \quad (1)$$

in which  $\alpha$  and  $\beta$  parameters can be estimated using MCNP simulations ( $\alpha = 0.64$  and  $\beta = 1.38$  for the reference sample) or measured experimentally with ore samples having different disequilibria. MCNP parametric studies have shown that the mineralogy (SiO<sub>2</sub> vs. Na<sub>2</sub>CO<sub>3</sub> or CaCO<sub>3</sub>) has a negligible impact on the spectrum, hence on  $\alpha$  and  $\beta$ , as well as the uranium grade up to 5000 ppm (beyond this value, gamma self-absorption by uranium becomes significant). However, when the density varies between 0.7 and 1.7 g.cm<sup>-3</sup>,  $\alpha$  and  $\beta$  vary respectively of  $\pm 1\%$  and  $\pm 12\%$  around their reference values (for 1.2 g.cm<sup>-3</sup>), and when the filling height changes between 1 and 7 cm,  $\alpha$  and  $\beta$  vary of  $\pm 2\%$  and  $\pm 19\%$  with respect to the reference filling height (4 cm). Therefore, the beaker will be systematically filled to a level of 4 cm ( $\pm 10\%$ ) and the density will be determined by weighing the crushed ore sample (with a precision of  $\pm 10\%$ ), which will limit the relative uncertainties on  $\alpha$  and  $\beta$  to about 1% and 7%, respectively. The disequilibrium  $D$  between U and Rn activities is deduced from (1):

$$D = \frac{A_U}{A_{Rn}} = \frac{C_{U \text{ band}} - \beta}{C_{Rn \text{ band}} \alpha} \quad (2)$$

And its relative standard deviation is:

$$\frac{\sigma_D}{D} = \sqrt{\left(\frac{\alpha \times D + \beta}{\alpha \times D}\right)^2 \times \frac{1}{\tau_U} + \frac{1}{\tau_{Rn}} + \left(\frac{\beta}{\alpha \times D}\right)^2 \times \left(\frac{\sigma_\beta}{\beta}\right)^2 + \left(\frac{\sigma_\alpha}{\alpha}\right)^2} \quad (3)$$

in which  $\tau_U$  and  $\tau_{Rn}$  are the count rates in the respective U and Rn bands (in s<sup>-1</sup>), and  $T_C$  is the counting time.

Figure 4 and Table I show that for a 300 s measurement, counting statistics may significantly affect the uncertainty on  $D$ , especially at low uranium grade or low U/Rn activity ratio.

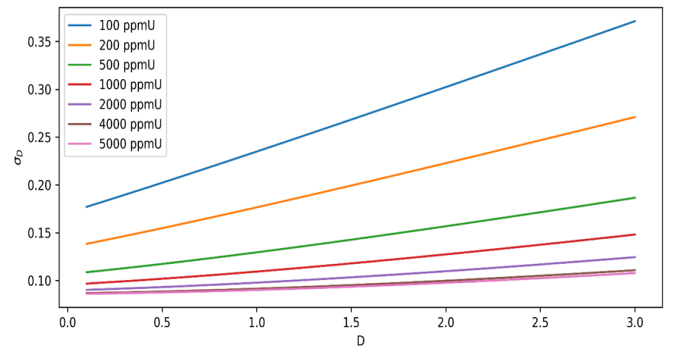


Fig. 4. Calculated standard deviation of  $D$  disequilibrium between U and Rn as a function of  $D$  and uranium grade, for a 300 s measurement time.

TABLE I  
EXAMPLES OF STANDARD DEVIATIONS AND RELATIVE STANDARD DEVIATIONS  
ON THE DISEQUILIBRIUM D BETWEEN U AND RN ACTIVITIES

U grade (ppm)	Disequilibrium D	$\sigma_D$	$\sigma_D/D$
100	0.2	0.23	120%
100	0.4	0.24	60%
100	1	0.27	27%
1000	0.2	0.16	80%
1000	0.4	0.17	35%
1000	1	0.17	17%

Figure 5 reports four spectra measured by ORANO Mining with real ore samples and Table II compares the results of the LaBr energy bands method with reference measurements of the uranium grade (ICP-MS) and U/Rn disequilibrium (HPGe). The overall agreement is satisfactory and the largest discrepancy is observed on the uranium concentration of sample 4. It could be due to local heterogeneities in the crushed ore and hence to the representativeness of the small sample extracted for ICP-MS analysis. Nevertheless, the 100 % numerical calibration performed with MCNP6.1 provides suitable results and more samples will be analyzed to confirm this preliminary conclusion.

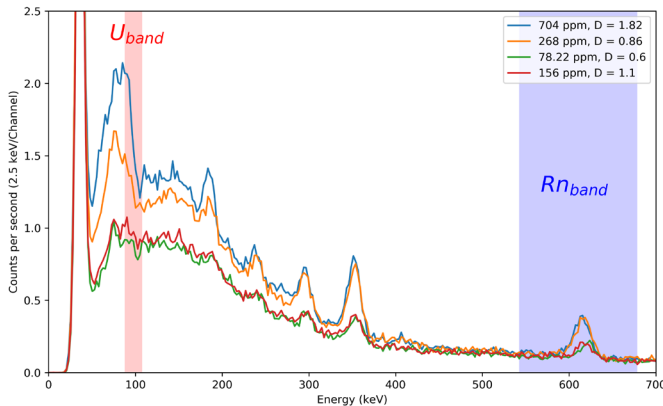


Fig. 5. Measured LaBr spectra of uranium ore samples.

TABLE II  
URANIUM GRADE AND U/RA DISEQUILIBRIUM MEASURED WITH LABR ENERGY BANDS, COMPARED TO REFERENCE HPGE OR ICP-MS MEASUREMENTS

Sample #	LaBr energy bands		Reference data	
	U/Ra	U grade (ppm)	U/Ra <sup>a</sup> (activity ratio)	U grade <sup>b</sup> (ppm)
1	2.53 ± 0.16	699 ± 44	1.82	704
2	0.73 ± 0.16	226 ± 48	0.86	268
3	0.71 ± 0.16	66 ± 15	0.6	78
4	1.06 ± 0.16	106 ± 16	1.1	156

<sup>a</sup> Measured by high resolution gamma spectroscopy (HPGe)

<sup>b</sup> Measured by ICP-MS

### III. LABR PROBE FOR BOREHOLE LOGGING

MCNP6.1 Monte Carlo simulations have been performed with the new LaBr probe to study the feasibility of the 1001 keV gamma-ray detection [4] in the calibration blocks of ORANO CIME (Innovation Center for Extractive Metallurgy) calibration station in Bessines, see Fig. 6, and of the two approaches based on low-energy energy bands [7]. This last is

illustrated in Fig. 7, with a narrow band (in blue) focused on the 98.4 keV self-fluorescence X-ray of uranium, induced by the gamma rays of the ore sample [1,2]. This band is insensitive to the uranium grade because of the competitive effects of self-absorption and self-fluorescence. However, the ratio of the counts in this band to that in the [560-660] keV band is linear with U/Rn disequilibrium, and same equations as (1) and (2) are used, here with  $\alpha_{\text{SXRF}}$  and  $\beta_{\text{SXRF}}$  coefficients (SXRF for self-induced X-ray fluorescence). Conversely, the low-energy band between 120 and 170 keV is inversely proportional to U grade up to 5000 ppm, see Fig. 8, while being insensitive to U/Rn disequilibrium D.

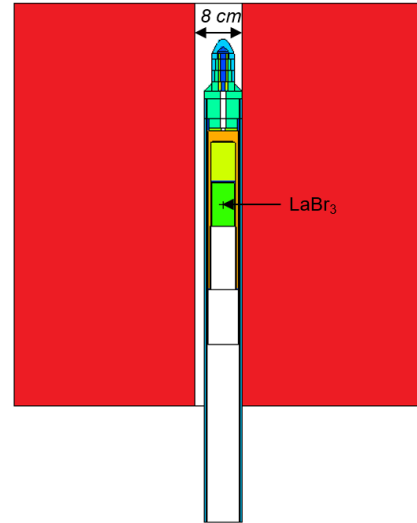


Fig. 6. MCNP model of the new LaBr probe in a reference concrete block of ORANO CIME calibration station (Bessines, France).

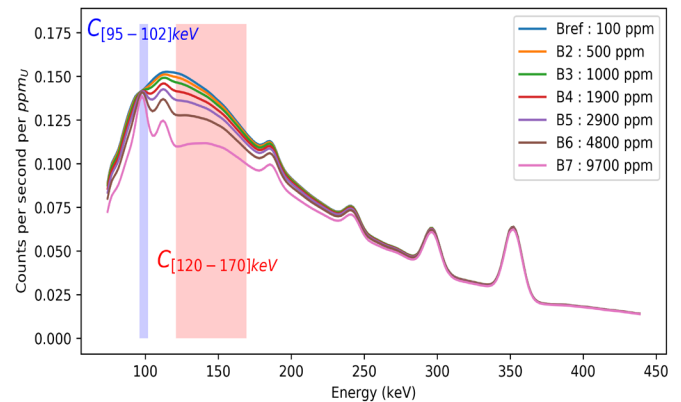


Fig. 7. MCNP6.1 simulated spectra of the LaBr borehole logging probe in the seven reference concrete blocks of ORANO CIME.

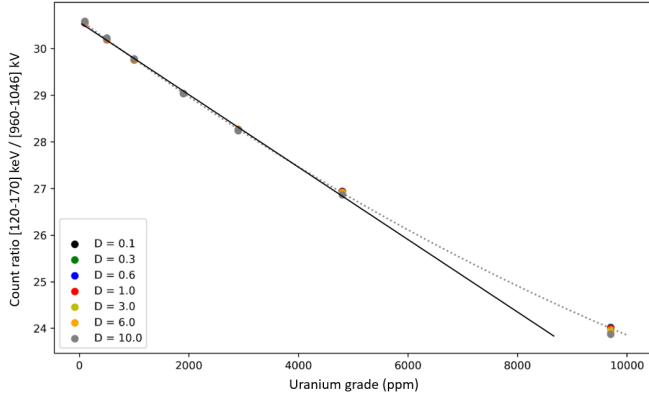


Fig. 8. Count ratio between the energy band from 120 to 170 keV (related to Compton continuum attenuation by uranium) and the band from 960 to 1046 keV (normalization), as a function uranium grade and disequilibrium.

The relationship between the count ratio and uranium mass concentration  $Cm_U$  (i.e. uranium grade in ppm) is written as:

$$\frac{C_{[120-170]keV}}{C_{[960-1046]keV}} = \beta_{Compton} - \alpha_{Compton} \times Cm_U \quad (3)$$

from which  $Cm_U$  is given by:

$$Cm_U = \frac{\beta_{Compton} - \frac{C_{[120-170]keV}}{C_{[960-1046]keV}}}{\alpha_{Compton}} \quad (4)$$

The LaBr probe was tested at ORANO CIME calibration station, in seven reference concrete blocks in radioactive

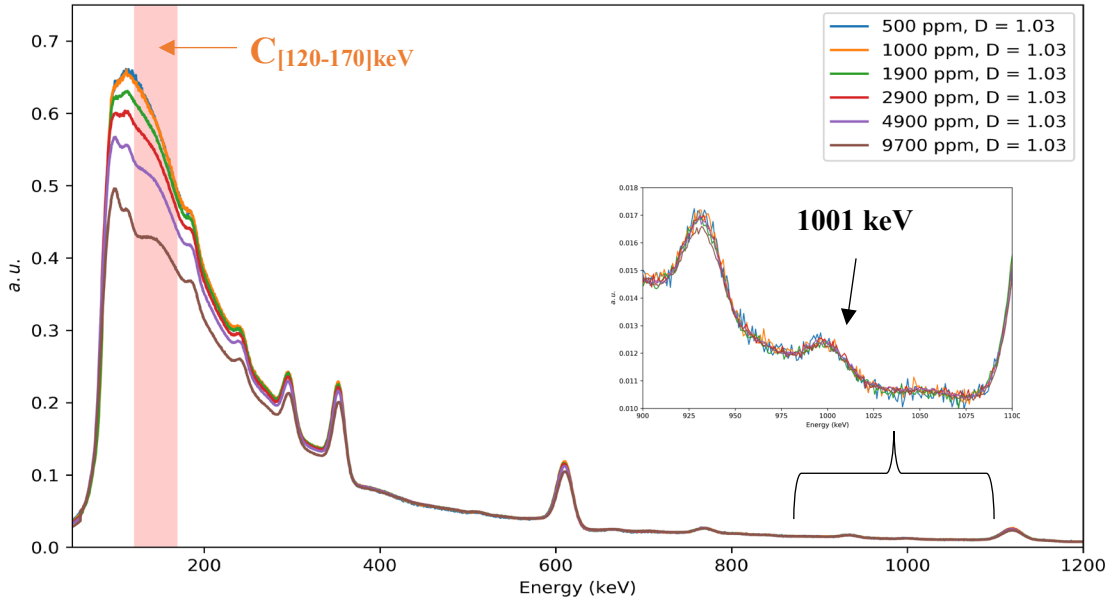


Fig. 9. Gamma spectra measured with the LaBr probe in the seven reference concrete blocks in radioactive equilibrium ( $D = 1.03$ )

equilibrium and with increasing uranium grades. The measured spectra are given in Fig. 9. These spectra are normalized with the counts of the band from 960 to 1046 keV. As shown in Fig. 10, the normalized number of counts in the low-energy band between 120 and 170 keV is inversely proportional to uranium grade (as in Fig. 8). Figure 10 also shows the linear relationship between the net area of the 1001 keV peak and the uranium concentration in each reference concrete block. The results of the approach using self-induced uranium fluorescence (disequilibrium) are not reported as the blocks are all in radioactive equilibrium ( $D = 1.03$ ).

#### IV. CONCLUSION AND PROSPECTS

After the preliminary experimental validation of the energy bands method with the new LaBr spectrometer, we will acquire more LaBr gamma spectra of uranium ore samples also characterized at ORANO CIME laboratories by HPGe and ICP-MS reference techniques, in order to complete the validation on a wider range of uranium grades and radioactive disequilibria. On the other hand, the LaBr probe will be sent in the coming weeks to the ISR (In Situ Recovery) mine of Katco, in Kazakhstan, to test the in situ calibration method [6] in reference boreholes. In addition, an optimization of its MCNP model will be carried out in order to study different effects such as density, borehole diameters or mineralogy. This optimization will be realized thanks to a comparison of experimental results and MCNP simulations for the measurements in the concrete blocks at ORANO CIME, in Bessines, France.

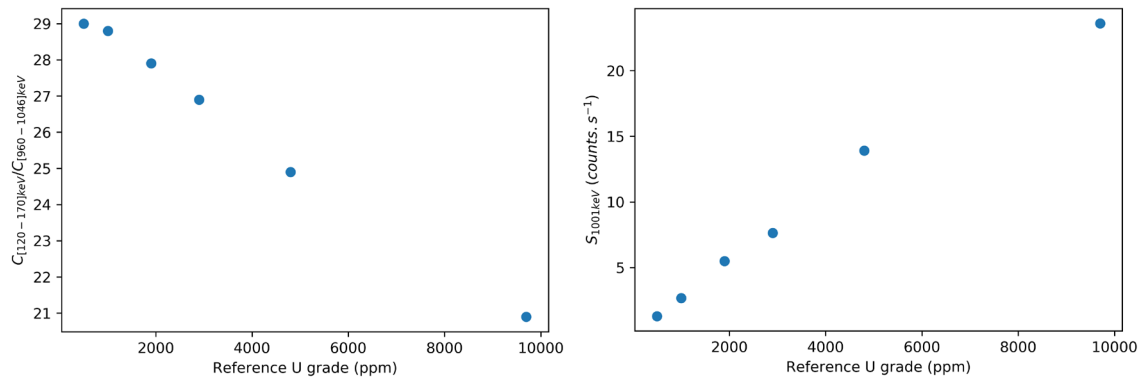


FIG. 10.  $C_{[120-170]keV}/C_{[960-1046]keV}$  RATIO AND  $S_{1001}$  COUNT RATE AS FUNCTION OF URANIUM GRADE OF ORANO CIME STANDARD CONCRETE BLOCKS

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