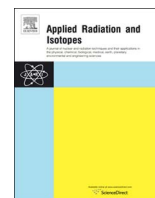




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## A gamma-ray spectrometry analysis software environment

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## HIGHLIGHTS

- A Monte Carlo code has been developed and validated using EGSnrc.
- A radionuclide decay generator has been written and validated.
- Detection efficiency can be calculated for High Purity Germanium detector.
- A flexible and user friendly analysis software using MS Excel® has been developed.
- Functionalities and performances of the software are presented.

## ARTICLE INFO

## Keywords:

Gamma-ray spectrometry  
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## ABSTRACT

At the JRC-Geel's RadioNuclide Metrology sector, a Monte Carlo code based on EGSnrc, and a general purpose calculation sheet implemented in Microsoft Excel®, have been developed to make the quantitative gamma-ray spectrometry analysis of samples simpler and more robust. The further aim is that the software can be used by non-experts in gamma-ray spectrometry e.g. external researchers using JRC-Geel's facilities through the EUFRAT transnational access scheme. This paper presents the developed Monte Carlo software and the functionality included in the calculation sheet.

## 1. Introduction

The JRC-Geel's RadioNuclide Metrology (RN) sector is operating low-background or ultra-low-background High Purity Germanium (HPGe) detectors. Currently, 11 of such detectors are operated in the underground laboratory HADES and 5 detectors are operated in the RN sector above ground laboratory at JRC-Geel. All detectors have distinct characteristics (type, size and manufacturer) as a wide range of samples with different geometries, masses or matrices have to be measured.

Since 2014, the facilities of the RN sector are accessible to external researchers through the transnational (or rather, "external", as also Belgian institutes can apply) access scheme called EUFRAT (European facility for nuclear reaction and decay data measurements) [<https://ec.europa.eu/jrc/en/eufrat>]. Up to date (March 2017), 22 projects have been approved by the external programme committee, for execution in the RN sector, 17 of which involve to some extent gamma-ray spectrometry.

The wide analytical capacity and large number of users of the RN sector gamma-ray spectrometry laboratories resulted in a clear need to employ a data analysis system that is robust and user-friendly so that external users can, following to a short introduction, perform their own

data analyses. A number of different approaches had been tested in the past. Although commercial computer codes for gamma-ray spectrometry are generally user friendly, it was found that they were not suitable for all types of samples/projects. Furthermore, it is of utmost importance for the RN sector to have full insight and control of the way the calculations are performed. This triggered the development of a general purpose MS Excel® based calculation sheet named "GLysis". "GLysis" offers the possibility to automatically incorporate spectra from Canberra Genie-2000 and Monte Carlo (MC) calculated Full Energy Peak (FEP) efficiency output data. An important aspect was to make the analysis robust by minimising copy/paste errors and need for manually entering data, while still maintaining flexibility. Another aspect of the design was to enable compliance with the ISO11929-2010 ISO standard. This paper describes this integrated system and discusses its performance and limitations.

Gamma-ray spectrometry using e.g. HPGe detector is a secondary method in the sense that the measurement of the activity in an unknown sample is conducted based on measurements of reference sources. The latter can be either activity standards or Certified Reference Materials (CRM). Availability of suitable reference sources is often limited. Computational methods using MC simulations have been

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**Table 1**

Comparison between the gamma-ray emission intensities generated by the decay generator and the DDEP data. The median values and the median absolute deviation (MAD) are calculated from the relative differences between the MC calculation and the DDEP. nb = number of particles/photons (beta-particles, gamma-rays and X-rays).

Nucl.	All			Only X-rays			Only gamma-rays ( $P_\gamma \leq 1\%$ )			Only gamma-rays ( $P_\gamma > 1\%$ )		
	nb	Median (%)	MAD (%)	nb	Median (%)	MAD (%)	nb	Median (%)	MAD (%)	nb	Median (%)	MAD (%)
<sup>227</sup> Ac	91	-98	2	4	-92.6	0.1	65	-98	2	0	-	-
<sup>228</sup> Ac	282	-3.3	3.2	4	-6.912	0.004	208	-2.2	2.2	17	0.001	0.079
<sup>133</sup> Ba	13	-0.003	0.092	4	0.086	0.001	2	-1.6	1.6	7	-0.7	0.1
<sup>214</sup> Bi	276	0.09	0.11	4	0.28	0.02	186	0.19	0.21	20	0.004	0.005
<sup>60</sup> Co	12	0.0005	0.1372	4	0.05	0.10	4	-3.77	3.93	2	0.0005	0.0005
<sup>134</sup> Cs	19	-0.002	0.019	4	0.003	0.012	3	-0.06	0.04	8	0.004	0.007
<sup>137</sup> Cs	9	-0.005	0.106	4	-0.112	0.002	1	0.24	-	1	0.0005	-
<sup>152</sup> Eu	132	-0.003	0.278	8	0.91	0.85	95	0.01	0.26	14	-0.01	0.02
<sup>234m</sup> Pa	145	-0.13	4.40	4	-96.63	0.01	119	-0.4	15.4	0	-	-
<sup>214</sup> Pb	30	-0.46	0.47	4	-0.918	0.003	15	-0.01	3.27	5	-0.001	0.006
<sup>223</sup> Ra	107	-0.60	2.16	4	-0.475	0.001	70	-0.6	0.3	7	-0.61	0.03
<sup>226</sup> Ra	14	0.02	0.09	4	-0.03	0.03	4	-0.2	0.4	1	-0.001	-
<sup>235</sup> U	77	-0.79	1.71	4	-1.71	0.01	44	-0.8	2.3	6	-3.4	3.0
<sup>212</sup> Bi	44	-0.27	62.96	4	-52.00	0.01	21	-0.07	0.16	4	-0.001	0.004
<sup>212</sup> Pb	13	0.0001	0.1509	4	-0.156	0.003	4	0.004	0.004	2	0.1	0.1
<sup>208</sup> Tl	46	0.14	0.21	4	0.07	0.01	21	0.14	0.05	6	0.1	0.1
All nucl.	1310	-0.073	0.465	68	-0.12	0.36	862	-0.07	83.25	100	0.00008	0.15000

proven to be a good alternative (albeit not as robust) to determine the FEP detection efficiencies. Even when a CRM is used to set up a FEP efficiency curve, in most cases, there is still the need to perform MC calculations to derive FEP corrections (efficiency transfer method) accounting for the (small) differences between the reference source and the analysed samples. At the RN sector calculations of FEP efficiencies and efficiency transfer factors are performed using the specially developed code "hpge3" using EGSnrc (Kawrakow et al., 2015). The "hpge3" code includes also a radionuclide decay generator, to account for true coincidence summing (TCS) effects.

## 2. The EGSnrc toolkit and "hpge3" code

EGSnrc is a general purpose simulation package including the coupled transport of electrons and photons. The approach of EGSnrc is different from a ready to use software. It provides a set of packages/classes written in C++ and a general purpose geometry library, which includes a set of particle sources and different propagation algorithms to simulate the interaction of the ionising particles. It is up to the users to write a programme for their specific needs in order to get the results for the quantities of interest (FEP efficiency in this case). The "hpge3" is based on the "EGS\_Advanced\_Application" C++ class (Kawrakow et al., 2017). Thanks to the EGSnrc C++ class library called "egspp", the model of the experimental setup and the definition of source(s) are defined in an input text file. No programming language and compilation are required to change setup. The structure of the input file is defined by.

Kawrakow et al. (2017). The EGSnrc toolkit does not include any radionuclide decay. Simulation of radionuclide decay is necessary in gamma-ray spectrometry to take into account the TCS effects which may occur when radionuclides with cascading gamma-rays are measured. For that purpose, a decay generator has been developed and included in "hpge3" software.

### 2.1. The decay generator

The decay generator is a C++ class which is added to the EGSnrc code but it can easily be included in other C++ MC codes. The class has been written in a way that allows new radionuclides to be added and nuclear decay data to be updated easily. The Decay Data Evaluation Project (DDEP) provides carefully evaluated decay data for frequently used radionuclides. The data are available in the ENSDF format (Tuli,

2001) and may be downloaded directly from the DDEP webpage (DDEP, 2004–2016). The decay generator uses such files as input. As not all radionuclides are available (e.g. <sup>227</sup>Th), the user may construct the missing nuclear data file, a simple ASCII file, following available instructions for its basic structure.

The decay generator is able to reproduce alpha, beta-, beta+ and EC decays. To simulate the decay the generator needs, as input, for each energy level, the energy of the level and the type of particle (alpha or beta- or beta+) with the associated energy. For simplification, emitted beta particles are not generated as a full beta spectrum but only at their mean energy. For each gamma-ray the following parameters must be provided: the gamma-ray energy, the emission probability ( $P_\gamma$ ), the  $K_\alpha$  and the total internal conversion factors ( $a_K$  and  $a_T$ , respectively). Only K-shell X-rays are simulated by providing their energy and emission probability and the fluorescence yield. The DDEP ENSDF data file does not provide the fluorescence yield. Thus, the fluorescence yields are written in a separate ASCII file.

Once all the decay data are read, the developed algorithm searches for all possible de-excitation paths to the ground state. For each path, a probability ( $P_{path}$ ) is calculated using the total probability for gamma transition including the conversion electrons ( $P_\beta$ ) of all the de-excitation gamma-rays. To simulate decay, a de-excitation path is selected randomly weighted by its  $P_{path}$  value. For each energy transition a random number is produced to generate either a conversion electron or a gamma-ray, according to  $a_T$ . In the case of a gamma-ray emission, a photon of the given energy is generated. If a conversion electron is selected, a second random number is generated to choose if the emitted electron is coming from the K-shell or not, depending on  $a_K$ . If a K-shell electron is selected, a third random value gives the energy of the electron according to the emission probabilities of the different K-shell electrons. Then the gamma-ray or the conversion electron is propagated until its energy is below the set threshold. For each transition in the selected de-excitation path, the same process is repeated. Once the ground state has been reached a new path is selected and a new event starts. If applicable, all possible radionuclide decay modes are simulated (in e.g. <sup>152</sup>Eu; <sup>152</sup>Eu disintegrates 72% to <sup>152</sup>Sm by EC and beta+ and 28% to <sup>152</sup>Gd by beta-). The simulations assume that the gamma-ray emissions are isotropic.

### 2.2. Decay generator performance

The performance of the decay generator was assessed by comparing the gamma-ray intensities derived from the simulated decay scheme

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