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Study on drum assay system setting for analysis of U-238 nuclide at KAERI

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1. Introduction

In order to evaluate the radionuclide inventory of the LL/ILW drums for the final repository, a drum assay system (Wide Range-Segmented Gamma Scanner; WR-SGS) have been operated since 2015. This system uses a non-destructive method and can measure gamma-emitting radionuclides contained in most LL/ILW generated at KAERI [1]. Stored LL/ILW drums contains various nuclides such as Co-60, Cs-137, U-238. Compare with destructive method, drum assay system is time-effective as well as cost-effective. On this paper, we describe how to set appropriate library of WR-SGS based on U-238 decay chain properties for assaying soil drums contaminated U-238 as non-destructive method.

2. Methods and Results

2.1 Outline of the analysis

The spectrum is analysed by the GammaVision engine and the library. It identifies the peaks in the spectrum and then checks to see if these match the position of the expected peaks in the library. It checks to see if any of the peaks are two peaks overlapping each other. It reports the peaks the peaks it has identified from the library and any peaks it has found but not identified.

This list of identified peaks is then used by SGSAssay to determine the content of the drum. It looks at each peak and if it has a high uncertainty it rejects the peak. It then looks at each nuclide in the list and checks which of the lines in the library have been found and using the expected relative strengths from the library makes a decision if the nuclide is present.

2.2 Process for adding a nuclide

a. Identify nuclide to be measured : U-238

b. Identify any daughters that may be secular equilibrium: Secular equilibrium can only occur in a radioactive decay chain if the half-life of the daughter radionuclide B is much shorter than the half-life of the parent radionuclide A. In such a situation, the decay rate of A, and hence the production rate of B, is approximately constant, because the half-life of A is very long compared to the timescales being considered.)

Table Ⅰ: Decay chain of U-238(uranium series)

Nuclides	Half-life	Nuclides	Half-life
U-238		U-238	
↓	4.4E9Y	↓	4.4E9Y
Th-234		Th-234	
↓	24.1d	↓	24.1d
Pa-234m			
↓	1.17m	Pa-234m	
Pa-234			
↓	6.7h	↓	1.17m
U-234		U-234	
↓	2.5E5Y	↓	2.5E5Y
Th-230		Th-230	
↓		↓	
…		…	

c. Identify the gamma rays emitted by the nuclide of interest and the secular equilibrium daughters. From a suitable reference compile a list of the gamma ray lines emitted by the nuclide of interest and all the daughters in secular equilibrium. For daughter nuclides where the branching ratio of the daughter is not 100% this ratio needs to be factored into the branching ratio of the gamma lines for this nuclide. For examples, U-238 have 2 lines, Th-234 have 15 lines and Pa-234 have 203 lines.

d. Select the gamma ray lines to be entered into the library: The gamma lines that should be considered should be those with the higher branching ratios. Gamma lines with low energies are less easy to measure due to the Compton down scatter from higher energy lines also the low energy region is more populated with lines from other nuclides.

Table Ⅱ: U-238 2 lines at 13, 48 keV

Energy (keV)	Branching ratio (%)	Notes
13	8.7	To low energy
48	7.5	To low energy

Table Ⅲ: Th-234 15 lines 3 with branching ratio >1%

Energy (keV)	Branching ratio (%)	Notes
63.3	3.9	To low energy
92.3	2.6	Lots of peaks region
92.8	3.0	Lots of peaks region

Table Ⅳ: Pa-234m 130 lines 4 with branching ratio >0.1%

Energy (keV)	Branching ratio (%)	Notes
94.7	0.1	Lots of peaks region
98.4	0.2	Lots of peaks region
766.6	0.2	
1001.03	0.6	

Table Ⅴ: Pa-234m 203 lines 6 with branching ratio >10%, Pa-234 ratio to U-238 0.16%

Energy (keV)	Branching ratio (%)		Notes
	Pa-234	To U-238	
94.7	15.5	0.025	Lots of peaks region
98.4	25.1	0.040	Lots of peaks region
131.3	20	0.032	Lots of peaks region
569.3	10.4	0.017	
883.3	12.0	0.019	
926.7	11.0	0.018	
946.0	20.0	0.032	

The effect of other expected nuclides should also be considered. If another nuclides has a peak very near one of these peaks then the two peak may overlap forming a doublet. Doublets are deconvoluted into their separate peaks by the GammaVision analysis engine but there is greater uncertainty on these results.

e. Setup the entry for the library: GammaVision Calculated activities are not used; peaks are processed directly by SGSAssay and thus it is not necessary to assign key line or Not-in-Average flags in the GammaVision library.

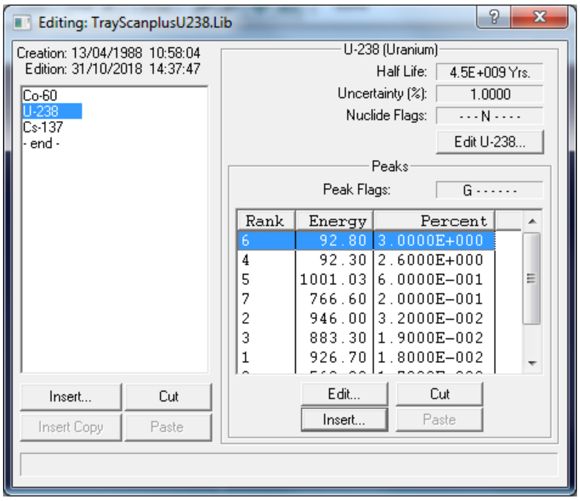
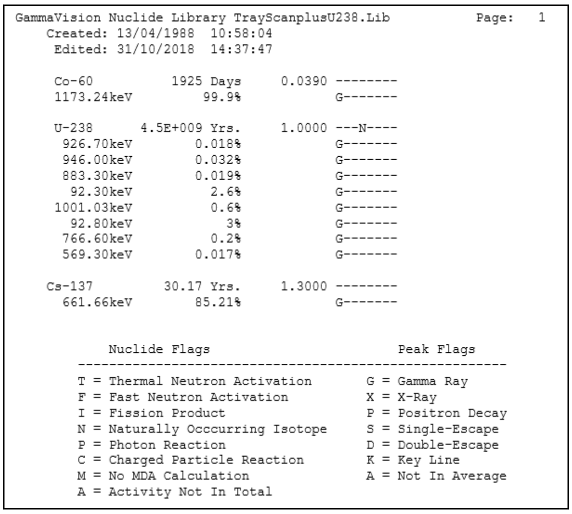


Fig. 1. GammaVision Nuclide library editor [2]



```
GammaVision Nuclide Library TrayScanplusU238.Lib          Page:   1
    Created: 13/04/1988  10:58:04
     Edited: 31/10/2018  14:37:47

     Co-60          1925 Days     0.0390 --------
     1173.24keV         99.9%            G-------

     U-238      4.5E+009 Yrs.     1.0000 ---N----
      926.70keV        0.018%            G-------
      946.00keV        0.032%            G-------
      883.30keV        0.019%            G-------
       92.30keV          2.6%            G-------
     1001.03keV          0.6%            G-------
       92.80keV            3%            G-------
      766.60keV          0.2%            G-------
      569.30keV        0.017%            G-------

    Cs-137         30.17 Yrs.     1.3000 --------
      661.66keV        85.21%            G-------


           Nuclide Flags                      Peak Flags
       -------------------------------------------------------
       T = Thermal Neutron Activation      G = Gamma Ray
       F = Fast Neutron Activation         X = X-Ray
       I = Fission Product                 P = Positron Decay
       N = Naturally Occcurring Isotope    S = Single-Escape
       P = Photon Reaction                 D = Double-Escape
       C = Charged Particle Reaction       K = Key Line
       M = No MDA Calculation              A = Not In Average
       A = Activity Not In Total
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Fig. 2. GammaVision Nuclide library report [2]

2.3 Further considerations

2.3.1 Number of peaks to include

Because the SGSAssay analysis will reject a nuclide if it does not detect a significant proportion of the gamma lines (this is weighted based on the branching ratio) so there is a greater chance of rejection measured gamma lines with more lines in the library.

There is also a risk with having a low number of lines in the library. The random scatter of the counts in each channel of the spectrum can lead to GammaVision detecting a peak with a low enough uncertainty to be accepted when there is none of the nuclide present. The more lines in the library for this nuclide the less likely this is to happen. The very low detection efficiency means that a few counts will be transformed into a few thousand or million Becquerels of activity.

The other side of this effect is that at the limit of detection the more lines in the library the less likely it is to give a measured result rather than an MDA.

2.3.2 Half-life limitations

If the half-life for a nuclide in the library is to short then the SGSAssay analysis engine will reject it since the activity changes to quickly. The activity of a nuclide with a half-life of 7 hours will change by more than 5% in half an hour (approximate duration of a measurement). This means daughter in secular equilibrium must be added under the parent as described below.

2.3.3 Using daughter gamma rays

It is common for the gamma lines of nuclides in secular equilibrium to be listed under the parent nuclide. The gamma lines from Cs-137 are in fact emitted by its daughter Ba137m which has a half-life of 153s. This allows the gamma rays from daughter nuclides with short half-lives to be used to quantify the parent. This can cause confusion when a gamma line is listed as coming from the parent when it is in fact emitted by the daughter.

3. Conclusions

Using drum assay system, analysis method of soil drums contaminated U-238 nuclide is developed as researching on decay chain of U-238(uranium series), properties of assay system and soil material properties. The analysis of several soil drums with the new setting library have a reliable value because of peak shape and count rates of measured spectrums. Many soil drums will be measured and the U-238 measuring system will be improved. To ensure quality of the results for U-238 nuclide, we plan to compare with those results from destructive method such as alpha spectrometry, ICP-MS (Inductively Coupled Plasma-Mass Spectrometry), neutron activation analysis [3].

REFERENCES

[1] W. H. Jang, K. K. Kwak, D. S. Hong, T. K. Kim and S. U. Jang, “Installation and Performance Test of a LL/ILW Drum Assay System at KAERI”, WM2014 Conference, Phoenix, Arizona, USA, March 2-6, 2014.

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[3] W. N. Lee, H. R. Kim, K. H. Chung, Y. H. Cho, M. J. Kang, C. W. Lee, G. S. Choi, “Uranium activity analysis of soil sample using HPGe gamma spectrometry”, Journal of radiation protection, Vol. 3 No.3 Sep, 2010