Radioisotope identification

Half-life and spectrometry

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Abstract

Research nuclear reactors such as the GSTR are capable of identifying the composition of unknown samples. This is done by bombarding them with neutrons to activate them, obtaining the half-life of the activation products and identify the gamma-ray discrete energies released. A library can then be used on the spectrum to identify which nucleides decay generated the various peaks.

This method can also give the quantities (mass) of each nucleide in the sample. However, only an energy calibration of the spectrometer was performed, hence this information was not made computed.

This report is organized as follow. In the first part, the theory is explained so that the objective of this experiment becomes clear. In the second part, the procedures used are described. Then, the results obtained are presented and error and uncertainties are finally discussed.

The library used during this experiment showed, in hindsight, a weakness. The main peak generated by the sample was not identified as it should have been. Ongoing work is being performed to update this library at the GSTR.

Table of Contents

Page

List of Tables

Table **Page**

LIST OF FIGURES

Chapter 1

Theory

Research nuclear reactors such as the GSTR are capable of identifying the composition of unknown samples. This is done by bombarding them with neutrons to activate them, obtaining the half-life of the activation products and identify the gamma-ray discrete energies released. A library can then be used on the spectrum to identify which nucleides decay generated the various peaks.

The procedure is issued from handouts from the USGS-Reactor Lab course at the Colorado School of Mines [2].

1.1 Radioisotope half-life

The neutron bombardment of samples at the GSTR facility activates the nucleides in the samples with an excess of neutron. The decay of those radioisotopes gives way to mostly β and γ.

Every isotope emits radiation with a constant decay rate and at discrete energies for gamma rays. Hence, the half-life – time after which half of the radioisotopes have decayed – and the gamma ray energies can be used to identify the radioisotope.

The decay of a radioisotope is given by:

$$A_f = A_0 e^{-\lambda t} \tag{1.1}$$

where:

A_f = Final activity
A_0 = Initial activity
λ = Decay constant
t = Decay time

The half-life can easily be determined from the data fit using equations 1.2 to 1.4:

$$\frac{A_0}{2} = A_0 e^{-\lambda t_{1/2}} \tag{1.2}$$

$$\ln(\frac{A_0}{2}) = \ln(A_0 e^{-\lambda t_{1/2}}) \tag{1.3}$$

$$t_{1/2} = \frac{\ln(2)}{\lambda} \tag{1.4}$$

After measuring the counts per set amount of time during a period of time covering at least a rough half-life estimate (activity divided by two), it is consequently trivial to fit the data to the exponential function described in equation 1.1, thus finding the decay constant λ and the half-life.

The R^2 value, representing the fit quality, can be found using the mean ($\bar{y}$), the total sum of squares (SS_{tot}), and the residual sum of squares (SS_{res}). Each is defined as:

$$\bar{y} = \frac{1}{n}\sum_{i=1}^{n} y_i \tag{1.5}$$

$$SS_{tot} = \sum_i (y_i - \bar{y})^2 \tag{1.6}$$

$$SS_{res} = \sum_i (y_i - f_i)^2 \tag{1.7}$$

$$R^2 = 1 - \frac{SS_{res}}{SS_{tot}} \tag{1.8}$$

where:

f_i = Exponential decay function value at point x_i

The decay constant and half life alone cannot be used to determine the isotopic composition of an unknown sample. Indeed, several activated isotopes are present and their resepctive activity and decay constants interfere. It can however narrow the possibilties. The spectrometry is then used to determine the different gamma ray emission energies and from that, determine the possible isotopes emitting such radiation.

1.2 Spectrometry

Most radioactive sources produce gamma rays, which are of various energies and intensities. When these emissions are detected and analyzed with a spectroscopy system, a gamma-ray energy spectrum can be produced. A detailed analysis of this spectrum is typically used to determine the identity and quantity of gamma emitters present in a gamma source, and is a necessary tool in a geochemical composition investigation. The gamma spectrum is characteristic of the gamma-emitting nuclides contained in the source. It is easy, once the emitting nuclides have been identified, to link them to their non-activated parent.

In order to get the most precise meausrement possible, an energy calibration must be performed. This consists of using a well-known sample to compare the measurements from the spectrometer to the expected gamma ray energies. The wanted precision is within 1keV, corresponding to the identification energy tolerance of the software used.

In a gamma-ray spectrometer there is a finite processing time required to measure and record each detected gamma ray, typically in the range of microseconds to tens of microseconds. During this processing time, called "dead time", the spectrometer is not able to respond to another gamma ray. This dead time implies that since gamma-ray photons arrive at the detector with a random distribution in time, some photons will not be measured or counted. The dead time should thus not exceed 10% in order to not lose too much information.

A software is used to process the data and remove gamma ray interference. A library is then used to link the measured peaks with emitting nuclides.

1.3 Procedure

This experiment has three components:

1. Irradiation of the sample
2. Half-life and decay constant determination
3. Sample spectrometry

1.3.1 Irradiation

In order to irradiate the sample, the reactor is set at full power. The sample (<0.01 g) is then lowered into a sample tube in the reactor bay, and is left within the neutron flux (approximately $1e^{12}n.cm^{-2}.s$ for a set period of time, between 10 seconds and five minutes.

The samples are then taken out of the reactor and brought to the lab for analysis.

1.3.2 Half-life and decay constant

The background radiation is measured, in order to substract it from the sample radiation, even though it is negligible and well within the measurement uncertainties.

The G-M (Geiger-Muller) detector is set up with the sample using an appropriate fixed geometry. The counts per second should not exceed 1500 to avoid the detector saturation. At chosen time intervals, the count data is recorded and plotted. When the count has been divided by two, half-life has been reached and no more data points are needed.

The data can then be fit to equation 1.1 and the half-life and decay constant can be found.

1.3.3 Spectrometry

The sample is then inserted inside a previously-calibrated using a common source (Europium at the GSTR facility). The gamma ray spectrum is then measured and analyzed automatically by the spectrometer software.

CHAPTER 2

Results

This chapter presents the results obtained during the experiment performed on September 14th, 2016. It presents the sample's half-life, its decay constant, and the analysis from the detector, allowing us to determine the isotopic composition.

2.1 Sample activity

The data, presented in appendix A, is plotted on figure 2.1, along with its exponential least-square fit. One can appreciate the good fit obtained, with an R^2 value of 99.5%, computed using equations 1.5 to 1.8.

The exponential decay of the sample follows:

$$A_f = 88877.6e^{-0.01926t} \tag{2.1}$$

Using the fitted value obtained for λ, equation 1.4 gives us the sample half-life $t_{1/2}$ at 35.98 minutes. This value is of no use to us in order to determine the isotopic composition of the sample.

2.2 Sample emissions

The spectrometer use was a little less straightforward. The library is apparently missing some radioisotopes and their associated gamma ray peaks, thus it could not find the elements emitting the primary peak. Moreover, the peak analysis report is missing quite crucial information about the actual elements associated to the peaks. The results with the peaks of interests (cut off at a net peak area of 1000) are given in appendix B.

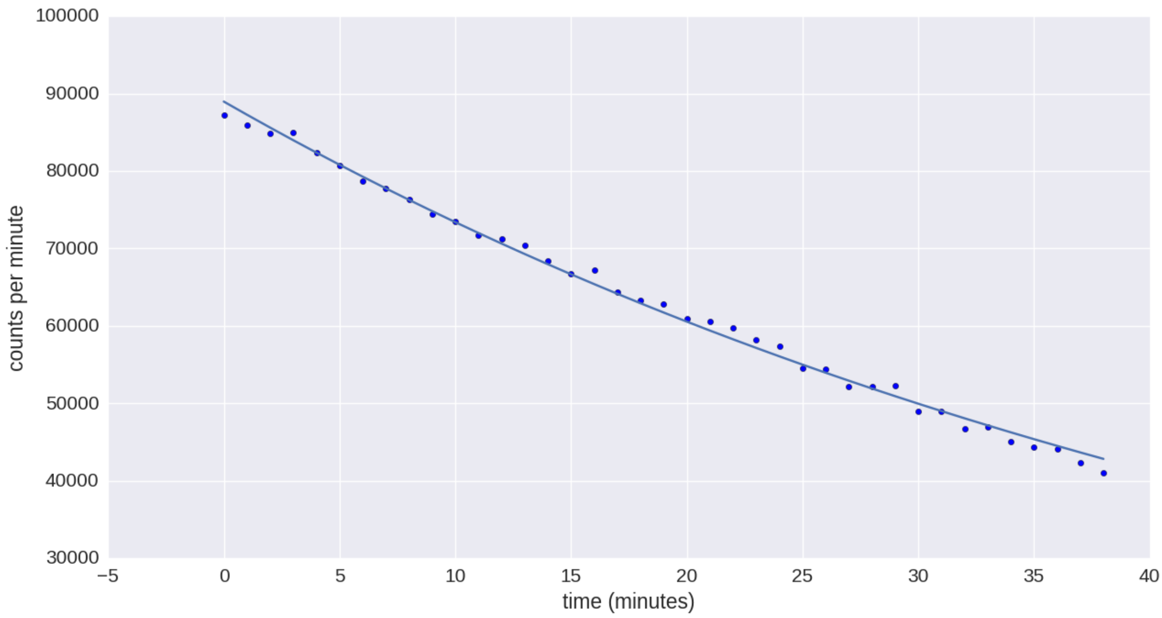

FIGURE 2.1. Activity of the sample.

Chapter 3

Conclusion

Appendix

A

Activity measurements

This appendix presents the activity measurements as a function of time. The activity was measured using a Geiger-Muller detector, over a 30 seconds period. In this table, the activities have been normalized to a minute interval. The activities have also been adjusted to the background noise, calculated at 89 counts per minutes.

Time (minutes)	Activity (counts per minute)
0	87188
1	85840
2	84734
3	84846
4	82272
5	80654
6	78664
7	77636
8	76254
9	74294
10	73382
11	71672
12	71086
13	70334
14	68244
15	66654
16	67124
17	64296
18	63216
19	62736
20	60786
21	60528
22	59676
23	58100
24	57236
25	54460
26	54314
27	52134
28	52110
29	52186
30	48856
31	48872
32	46594
33	46860
34	44926
35	44220
36	44018
37	42226
38	40920

Table A.1: Activity measurements

Appendix B

Detailed data tables

This appendix presents the data measured during the spectrometry. Given the extensive peak analysis report automatically done by the spectrometer software, only the peaks presenting a net peak area greater than a thousand have been considered. The most likely elements, based on the gamma ray peak energy and average half-life on the sample of around 36 minutes, have been selected. In this regards, for example, potential parent candidates with a half-life of less than a minutes were discarded, their significant presence one hour after irradiation being ruled out. In the same vein, potential parent candidates with a half-life of more than a year were also discarded in favor of shorter lived isotopes.

The data used comes from the NDS (Nuclear Data Services) department of the IAEA [1].

Peak number	Energy (keV)	Net Peak Area	Possible parent
2	86.52	7888.57	$^{155}_{65}Tb_{90}$
4	92.88	2639.50	$^{67}_{29}Cu_{38}$, $^{178m}_{71}Lu_{107}$, $^{178m}_{73}Ta_{105}$, $^{180m}_{72}Hf_{108}$
5	94.88	4650.38	$^{172}_{73}Ta_{99}$
8	108.20	1322.32	$^{105}_{43}Tc_{62}$, $^{131m1}_{56}Ba_{75}$, $^{131}_{57}La_{74}$, $^{137}_{61}Pm_{76}$, $^{151}_{65}Tb_{86}$
13	162.43	1132.79	$^{190}_{74}W_{116}$, $^{244}_{93}Np_{151}$
14	169.15	1613.15	$^{182}_{75}Re_{107}$, $^{124}_{56}Ba_{68}$, $^{164}_{65}Tb_{99}$
27	311.77	1583.95	$^{133}_{52}Te_{81}$, $^{173}_{72}Hf_{101}$, $^{177m2}_{72}Hf_{105}$
40	459.14	3313.21	$^{183}_{72}Hf_{111}$
56	669.74	1186.98	$^{205}_{85}At_{120}$

Table B.1: Spectrometry data

Bibliography

[1] Nuclear Data Services – IAEA, *ENSDF database maintained by the International Nuclear Structure and Decay Data Network*.
`https://www-nds.iaea.org/relnsd/NdsEnsdf/QueryForm.html`.
Accessed: 2016-09-15.

[2] T. Debey, *USGS reactor lab: Radioiostope identification*.
Handouts.
Accessed: 2016-09-14.