FOPRA - 61 FRM PGAA

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1 Introduction

The goal of this internship is to gain an insight into the world of neutron research and in this course to determine the major components of a cement sample using the PGAA experiment.

2 Theory

When a neutron is captured by a nucleus ${}^{A}_{Z}\mathrm{X}$, the compound nucleus ${}^{A+1}_{Z}\mathrm{X}^{*}$ is being formed. The compound nucleus is in an excited state with an excitation energy that equals the binding energy of the neutron. However, the excited state is not stable and decays in less than $10^{-10}\,\mathrm{s}$. The radiation that is emitted in this process is called **prompt gamma radiation**. The energies of the photons correspond to the energy levels of the nuclide and are thus characteristic to this nuclide. In most cases (except for H and D), the energy emission takes place in form of an energy cascade, leading to multiple characteristic photons being emitted by single one nuclide. If the de-excited nucleus is radioactive, it typically decays with β-decay which also leads to the emission of photons called **delayed gamma radiation**. These photons are also characteristic and the delay and the count rate are dependent of the nucludies half life.

In **prompt gamma activation analysis**, the characteristics of prompt gamma radiation can be used to determine the components of a sample.

The characteristic photons lead to gaussian peaks at the nucdlides characteristic energies in the spectrum. The amount of the element in the sample can then be identified with the **absolute method**:

$$\frac{A}{\epsilon \cdot t} = a \cdot P_\gamma = n\,\Phi_0\,\Theta\,\sigma_0\,P_\gamma = \frac{m}{M}\,N_A\,\Phi_0\,\sigma_\gamma, \tag{1}$$

where A is the peak area, ϵ the counting efficiency, t the measurement time, a the activity and P_γ the emission probability.
The absolute method can not be used if we do not know the actual mass of the sample or if we can not be sure that the beam flux actually reaches the sample. In that case, we can still use the **relative method**:

$$\frac{A_1/\epsilon_1}{A_2/\epsilon_2} = \frac{n_1\,\sigma_{\gamma,1}}{n_2\,\sigma_{\gamma,2}}. \tag{2}$$

The composition can then be calculated in weight or molar percent from the mass or molar ratios respectively.

This experiment uses a high-purity germanium semiconductor detector for the detection of photons. It is basically a large diode that only lets current flow when a electron-hole pair is created by a photon. The signal is then amplified and a digital spectrometer determines the signal height.

3 Procedure and Results

3.1 Task 1: Efficiency of the detector

Before using the detector, some preliminary tasks have to be performed: First, we need to calibrate the detector using a sample with accurately known photon energies. Since the detector at at the facility is 330 mm away from the sample, not all photons reach the detector. Therefor, we need to know the detector efficiency: $\epsilon = \frac{\gamma_{\text{detected}}}{\gamma_{\text{emitted}}}$. Using formula (1) we can derive an equation for the efficiency

$$\epsilon = \frac{A/t}{a \cdot P_\gamma}, \tag{3}$$

that can be calculated and put into a diagram together with the respective energies to fit a curve to it in order to get the efficiency function $\epsilon(E)$. The detector efficiency function is determined using a ${}^{152}\mathrm{Eu}$ source with a known emission probability for each characteristic energy an activity of 393,0 kBq $\pm 1.5\%$ at 1^{st} of March, 2008 and a half-life of 13.516 y. The energy values and their corresponding counting rates can be found by evaluating the emission spectrum of the ${}^{152}\mathrm{Eu}$ source. The peaks of

the spectrum can then be fitted with several programs that determine the net peak area or the count rate at those specific energies.

The efficiency at each given energy can be calculated with equation (3) which is derived from the absolute method (1). The count rate (number of gamma photons detected in a second) was obtained from analyzing the peaks in the spectrum and the emission probability was obtained from the spreadsheet that was provided in this experiment. The activity of the sample at the time of the experiment can be determined with:

$$a(t) = a_0 \cdot e^{-\lambda \cdot t} \qquad \text{with} \qquad \lambda = \frac{\ln(2)}{T_{1/2}}, \tag{4}$$

where $a_0 = 393{,}0\,\text{kBq}$ (2008-03-01), $T_{1/2} = 13{,}516\,\text{a}$ and t is the time between the measurement of a_0 and the experiment in years. Using that formula an activity of 224,0 kBq was found. For all the peaks that were found in the spectrum, the efficiency was calculated and plotted against the corresponding energy.

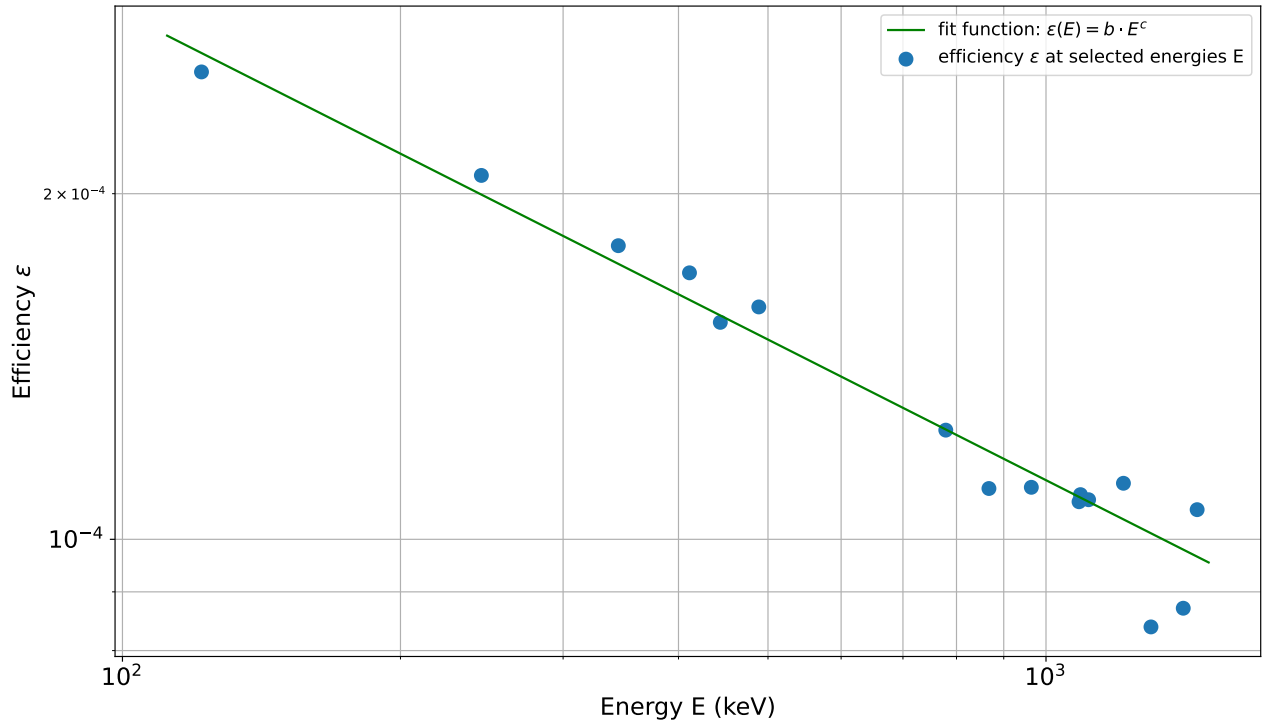

Figure 1: Efficiency of the detector at selected energies in log-log scale with fit function

Figure 1 shows the efficiency vs. energy for the ^{152}Eu sample, from which we determine the energy dependent efficiency of the detector using a straight line as a fit function:

$$\epsilon(E) = b \cdot E^c \tag{5}$$

with fit parameters $b = (1{,}87 \pm 0{,}20) \cdot 10^{-3}\,1/(\text{keV})^{\text{b}}$ and $c = -0{,}407 \pm 0{,}018$. It takes an energy in keV and returns the corresponding detector efficiency at that energy.

3.2 Task 2: Thermal-equivalent neutron flux

We also need to know the thermal-equivalent neutron flux Φ_0. It can be determined using a titanium flux-monitor foil. Φ_0 can then be calculated with:

$$\Phi_0 = \frac{A/t \cdot M}{\epsilon \cdot N_A \cdot \sigma_\gamma \cdot m}, \tag{6}$$

where N_A is the Avogadro constant, A/t the count rate, m the mass of the sample, M the molar mass and σ_γ the cross section of the sample. Using formula 6 we obtain $\Phi_0(341\,\text{keV}) = (2{,}092 \pm 0{,}315)\,1/\text{cm}^2\text{s}$ and $\Phi_0(1381\,\text{keV}) = (1{,}747 \pm 0{,}297)\,1/\text{cm}^2\text{s}$. The uncertainties are very high and come from the large uncertainties of the efficiency fit function, but the values themselves have the right order of magnitude when compared to the value given in the instructions ($3{,}0 \cdot 10^9\,1/\text{cm}^2\text{s}$ [1]). The two obtained values for 341 keV and 1381 keV should in theory be same, the difference comes once again from the uncertainties of the efficiency fit function.

3.3 Task 3: Partial gamma-ray production cross section of potassium

The next task was to determine the partial gamma-ray production cross section for 770-keV line of potassium from the PGAA spectrum of potassium chloride (KCl) using the following formula derived from the relative method (equation 2).

$$\sigma_{\gamma,2} = \frac{n_1\,\sigma_{\gamma,1} \cdot A_2/\epsilon_2}{n_2 \cdot A_1/\epsilon_1} \overset{n_1=n_2}{=} \frac{\sigma_{\gamma,1} \cdot A_2/\epsilon_2}{A_1/\epsilon_1} \tag{7}$$

The partial gamma-ray production cross section of chloride (517 keV) was given in the instruction manual of this experiment [1] as $\sigma_{\gamma,1} = 7{,}58\,\text{b} \pm 0.6\%$. The count rates were determined from the spectrum as described in section 3.1 and the efficiencies calculated with formula (5).

Table 1: Count rates and efficiencies for KCl (K: 770 keV, Cl: 517 keV)

element	count rate	efficiency
K	7.952	$0{,}000\,150 \pm 0{,}000\,155$
Cl	78.94	$0{,}000\,126 \pm 0{,}000\,134$

With the values from table (1) and the given cross section for the Cl line the value for the partial gamma-ray production cross section of the 770 keV line of potassium was calculated: $\sigma_{\gamma,2} = (0{,}90 \pm 0{,}20)\,\text{b}$ Although the uncertainty of the result is very high, it is very close to the value of the partial gamma-ray production cross section of the 770 keV line given in the excel files ($\sigma_{\gamma,Cl} = 0{,}903\,\text{b} \pm 1.6\%$ [1]).
The efficiency ratio is $\frac{\epsilon(517)}{\epsilon(770)} = 1.18$, which is very close to the efficiency ratio in February 2019 ($\epsilon(517)/\epsilon(770) = 1.21$ [1]).

3.4 Task 4: Composition of a cement sample

Next we want to determine the composition of a cement sample using a spectrum obtained with PGAA. The components of the sample can be identified by getting the energy of a peak in the spectrum and finding the element that emits photons with this energy [1]. One can then calculate the mass m of the portion of the element that is exposed to the neutron beam using:

$$m = \frac{A/t \cdot M}{\epsilon N_A \Phi_0 \sigma_\gamma}, \tag{8}$$

which is equation 1 solved for m.

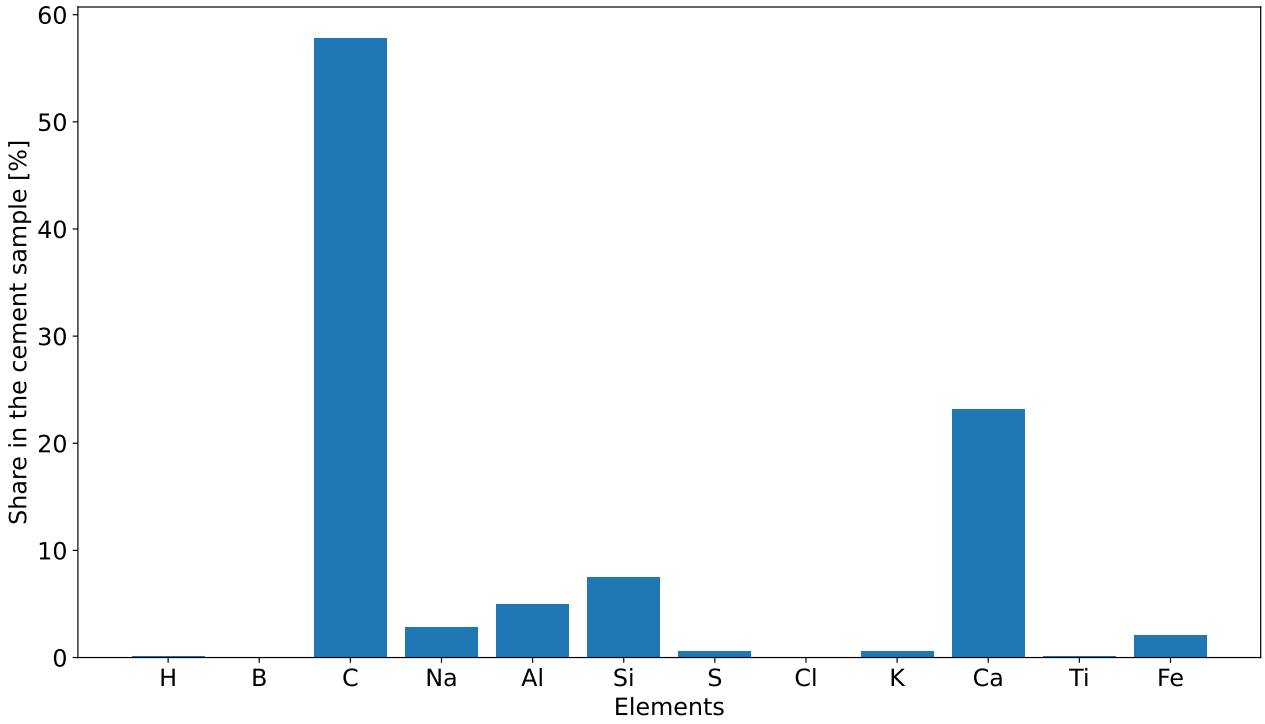

Figure 2: Composition of the cement sample without oxygen. C and Ca are the major components but it also contains other elements that can be found in the earth like silicon, aluminium, natrium and iron.

Figure 2 shows the composition of the cement sample and table 2 in the appendix the masses and mass percentages of selected elements. One can see that the cement sample mostly consists of carbon and calcium, which seems correct since limestone ($CaCO_3$) is a major ingredient of cement. It also contains elements commonly found in the earth like silicon and aluminium. Although oxygen is also a major component of cement, we could not measure its portion due to its small cross section. We also disregarded that a portion of the carbon peaks in the spectrum came from the teflon bag which held the sample.

A Appendix

Table 2: Masses of the elements

Element	m in μg	percentage
H	46.14,	0.16
B	1.25,	0.00
C	16383.	57.82
Na	810.8	2.86
Al	1413.	4.98
Si	2124.	7.49
S	181.75	0.64
Cl	9.71,	0.03
K	161.9,	0.57
Ca	6561.	23.16
Ti	52.74	0.18
Fe	584.4	2.06
total	28331	100

References

[1] Technische Universität München. Prompt gamme activation analysis (pgaa), FOPRA / Advanced Lab Course.