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# Preparational work for a measurement of the $^{146}\text{Sm}$ $\alpha$ -decay

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## Summary

### Abstract

The isotope  $^{146}\text{Sm}$  is a now entirely decayed nuclide that gives information about the chronology of the solar system. The half life of  $^{146}\text{Sm}$  was measured 5 times by different experiments, gaining values ranging from  $6.8 \cdot 10^7$  years to  $1.03 \cdot 10^8$  years. Because of the importance of this half life information, it will be remeasured with a significant number of decays to get a reliable value. This work concentrates on calculations and problem solving that come along with the production of  $^{146}\text{Sm}$ . Due to a gamma spectroscopy done within the scope of this thesis, a limit for the half life of the first excited state of  $^{146}\text{Nd}$  was determined.

### Zusammenfassung

Das inzwischen vollständig zerfallene Isotop  $^{146}\text{Sm}$  gibt Aufschluss über das Alter und die Chronologie des Sonnensystems. Die Halbwertszeit dieses Nuklids wurde bisher 5 mal im Rahmen verschiedener Experimente gemessen, jedoch reichen die gemessenen Halbwertszeit von  $6.8 \cdot 10^7$  Jahren bis  $1.03 \cdot 10^8$  Jahren. Daher soll die Halbwertszeit mit Augenmerk auf eine ausreichenden Zahl an Zerfällen erneut gemessen werden, um einen vertrauenswürdigen Wert für die Halbwertszeit zu erhalten. Diese Arbeit beschäftigt sich mit Berechnungen und Problembetrachtungen, die mit der Herstellung des natürlich nicht mehr vorhandenen Isotops  $^{146}\text{Sm}$  einhergehen. Im Rahmen dieser Arbeit wurde eine Gammaspektrometrie durchgeführt, bei deren Auswertung ein Limit für den ersten angeregten Zustand von  $^{146}\text{Nd}$  bestimmt wurde.



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# 1 Introduction

Even in the year 2015 the age of the universe is an ongoing question. The  $\alpha$ -decay of the now extinct nuclide  $^{146}\text{Sm}$  into  $^{142}\text{Nd}$  brings conclusions to the solar chronology. In the last 60 years the half life of  $^{146}\text{Sm}$  has been measured several times, but with up to a 30 % difference between the single half lives and to the tabular value of  $1.03 \cdot 10^8$  years, compare Table 1. Only the values from Friedman et al.(1966) and Meissner et al.(1987) [4, 5] were used to calculate the tabular value.

Year	Half life in years	Reference
1963	$8.5 \times 10^7$	Friedman et al. [1]
1964	$7.4 \times 10^7$	Nurmia et al. [13]
1966	$1.03 \times 10^8$	Friedman et al. [4]
1987	$1.03 \times 10^8$	Meissner et al. [5]
2012	$6.8 \times 10^7$	Kinoshita et al.[14]

Table 1: Half life measurements of  $^{146}\text{Sm}$ .

Therefore, the half life of  $^{146}\text{Sm}$  should be remeasured with attention on activity and measuring time to have a significant differentiation between the listed measurements, which are calculated within this work.

Due to the extinction of  $^{146}\text{Sm}$  it has to be produced via proton activation of  $^{146}\text{Nd}$  and the  $\beta^-$ -decay of  $^{146}\text{Pm}$ . The starting point for this process is a sample of 100 mg  $\text{Nd}_2\text{O}_3$  powder.

Considering the production of  $^{146}\text{Sm}$  preparational work and calculations as well as looking at possible problems during activation has been done in Chapter 3 and 4 . Within the analysis of a gamma spectrum for the  $\text{Nd}_2\text{O}_3$  sample, a limit for the half life of the first excited state of  $^{146}\text{Nd}$  has been determined in Chapter 4.3. This  $\alpha$ -decay to  $^{142}\text{Ce}$  has been never observed before.



## 2 Fundamentals

Before starting with calculations for the half life of  $^{146}\text{Sm}$ , the experimental schedule as well as fundamental terms will be explained. Possible problems occurring during activation will be listed and discussed later on.

### 2.1 Experimental Schedule

A rough schedule for the whole procedure is shown in Figure 1. The starting point is a sample of Neodymium-oxide ( $\text{Nd}_2\text{O}_3$ ) powder. The mass of this sample is 100mg and has an abundance of  $97.2\% \pm 0.1\%$  of  $^{146}\text{Nd}$  in contrast to an abundance of about  $17.2\%$  in natural Neodymium.

This sample will be activated via proton bombardment and  $^{146}\text{Pm}$  is obtained.  $^{146}\text{Pm}$  decays via electron capture into  $^{146}\text{Nd}$  and via  $\beta$ -decay into  $^{146}\text{Sm}$ .  $^{146}\text{Sm}$  decays via  $\alpha$ -decay into  $^{142}\text{Nd}$  and the half life of this decay should be remeasured.

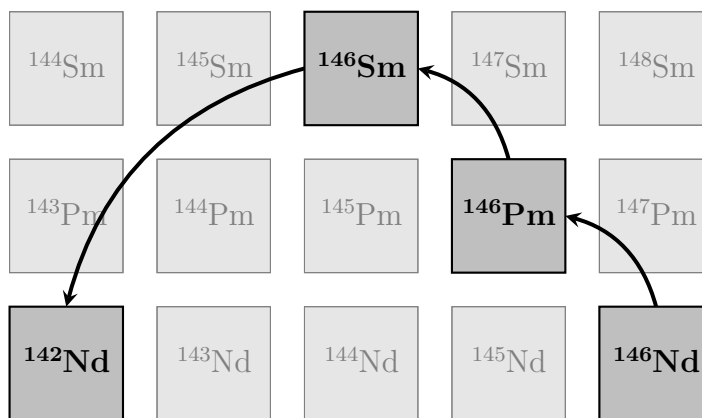


Figure 1: Schematic Schedule of measuring the half life of  $^{146}\text{Sm}$ .

### 2.2 Interaction of Alpha Particles

Looking at interactions between particles and matter, it can be distinguished between direct and indirect ionizing radiation. Alpha particles participate in direct ionization like all charged particles. There are different ways of ionizing directly: There can be inelastic collisions with the Coulomb field of the nucleus. The particle is slowed and bremsstrahlung occurs. This effect is relevant for high particle energies [10].

Other possibilities are collisions with shell electrons. On the one hand there can be elastic scattering, where the particle loses nearly no energy. On the other hand inelastic scattering can occur, where the atom is ionized or excited. Elastic scattering is only of interest at low particle energies.

For alpha particles the main interaction for energy transfer to the detector is the ionization of the atom because of the double positive charge of the alpha particles.

### 2.3 Alpha Chamber

The detector used here is an alpha chamber, which is based on a grid ionization chamber. It is filled with P10 gas, a gas mixture of 90% argon and 10% methane.

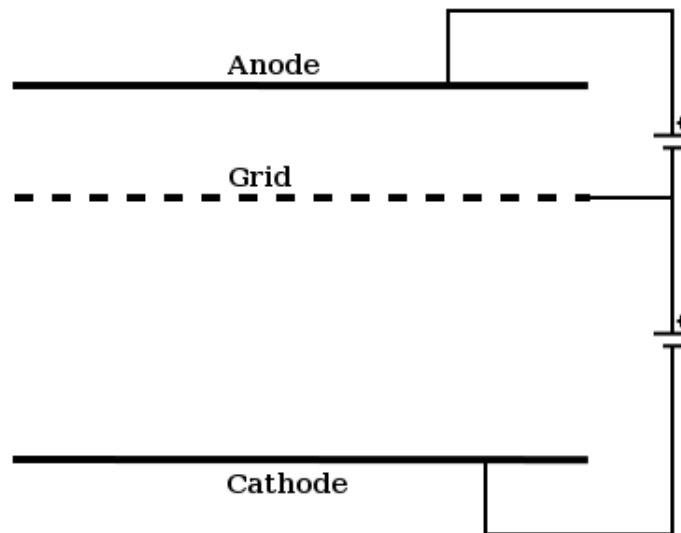


Figure 2: Structure of a grid ionization chamber

In the chamber there is an electric field between anode and cathode. After an alpha decay the alpha particle interacts with the gas molecules and ionizes them. The ionized particles move towards the electrodes due to the electrical field and can be measured. The structure of a grid ionization chamber can be seen in Figure 2.

To improve the efficiency of the chamber, nearly no oxygen should remain in the chamber, because oxygen captures free electrons that are needed for the measurement and the signal is reduced. Therefore the chamber is flushed with the P10 gas multiple times, because argon has a low electron affinity.

### 2.4 Fundamental Terms

#### Activity and decay constant

Dealing with decays always leads to a certain *activity*  $A$ . Activity can be described as the time derivation of the *number of decays*  $N$ . Besides the derivation relation between these two, it is also possible to describe the relation with a factor  $\lambda$ , known as *decay constant*, which is defined as  $\lambda = \frac{\ln 2}{T_{1/2}}$  with the half life  $T_{1/2}$ :

$$A(t) = -\frac{dN(t)}{dt} = \lambda N(t) \quad (1)$$

### Cross Section

The *cross section* is used to make a prediction about the probability a reaction is performed. It is defined as the mean *number of interactions with the target*  $N$  by the *particle fluence*  $\Phi$ , the target is exposed:

$$\sigma = \frac{N}{\Phi} \quad (2)$$

That means, the higher the cross section the more interactions take place. The cross section is energy dependent. An example for energy dependency of the cross section of  $^{146}\text{Pm}$  produced via proton bombardement of  $^{\text{nat}}\text{Nd}$  can be seen later on in Chapter 4.1.

### Particle Fluence and Flux

The *particle fluence* is defined as the number of interacting particles divided by the area  $A_{\perp}$  of the target that is perpendicular to the particle direction:

$$\Phi = \frac{N}{A_{\perp}} \quad (3)$$

The time derivation of the fluence  $\Phi$  is called *flux*. Knowing the used current  $I$  and the charge of the activating particles  $Q$  it can be calculated by

$$\varphi = \frac{I}{Q \cdot A_{\perp}} \quad (4)$$

### Q-Value

The energy gain of a reaction can be described by the Q-value, which is the difference between the energy that exists before the reaction and the energy after the reaction. If the Q-value is positive, energy is released.

## 2.5 Activation

The first step in producing  $^{146}\text{Sm}$  is the activation of the given sample. Therefore the (p,n)-reaction and possible problems occuring with the activation are discribed in the following.

**(p,n)-reaction**

To get  $^{146}\text{Pm}$ , the  $^{146}\text{Nd}$  sample is activated with protons. After bombarding a neutron is emitted and  $^{146}\text{Pm}$  results. The process for this activation can be described as follows:

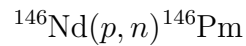


Figure 3 illustrates this reaction. Protons are accelerated with certain energy. These protons hit the target ( $^{146}\text{Nd}$ ), each one setting free a neutron and producing  $^{146}\text{Pm}$ .

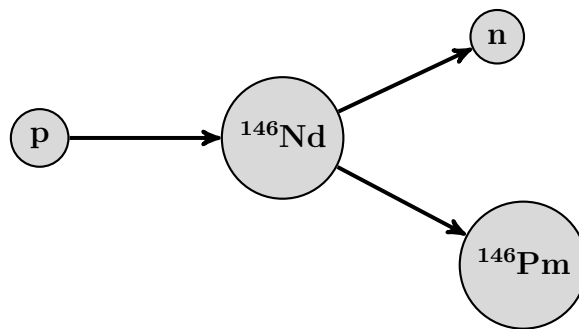
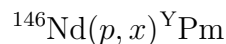


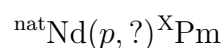
Figure 3: Schematic view of a (p,n)-reaction

**Problems with Activation**

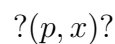
Activating the  $\text{Nd}_2\text{O}_3$  sample can cause three main problems, all can lead to other produced nuclides, which can decay with a much shorter half-life and with  $\beta$ -decays. Different possibilities and their probabilities have to be concerned. First another nuclide of Pm can be obtained after bombarding  $^{146}\text{Nd}$  with a proton. For different energy ranges there are various reactions, e.g. (p,n), (p,3n):



Second because of an abundance of 97.2% for  $^{146}\text{Nd}$ , the assumption is made that there are about 2.8% other Nd isotopes in the target and are considered as  $^{\text{nat}}\text{Nd}$ .



The last problem results from any other contaminations that might be in the sample:



In chapter 4.2 it will be discussed, how to handle these problems.

## 2.6 Decays

During preparation and measurements, different decays occur [9]. First there is  $\beta$  - decay and electron capture (EC) for the decay of  $^{146}\text{Pm}$ . Second, there is an  $\alpha$ -decay from  $^{146}\text{Sm}$  to  $^{142}\text{Nd}$ .

### $\beta$ -decay

The produced  $^{146}\text{Pm}$  has two decay modes. 66% of it decays into  $^{146}\text{Nd}$  via a  $\beta^-$ -decay. 34% decays via EC into  $^{146}\text{Sm}$  [8, 7].

The nuclear reaction for the  $\beta^-$ -decay is

$$n \rightarrow p + e^- + \bar{\nu}_e$$

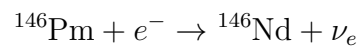
and in case of  $^{146}\text{Pm}$ :



Electron capture is described by

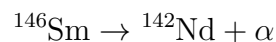
$$p + e^- \rightarrow n + \nu_e$$

and for this example



### $\alpha$ -decay

$^{146}\text{Sm}$  decays with an  $\alpha$ -decay into  $^{142}\text{Nd}$ .



The emitted  $\alpha$ -particle later on interacts with the gas molecules and the ionized molecules are detected by the alpha chamber.

### $\gamma$ -Ray Emission from Excited States

IT stands for the  $\gamma$ -rays, which are emitted from short-lived excited states that fall back to the ground state.

### Decay law

For the decay of  $^{146}\text{Pm}$  the well-known decay law can be used:

$$N_1(t) = N_1(0) \cdot e^{-\lambda_1 t}, \quad (5)$$

where  $\lambda = \frac{\ln 2}{T_{1/2}}$ .

Looking at the  $\alpha$ -decay of  $^{146}\text{Sm}$ , the production of  $^{146}\text{Sm}$  from the mother nuclide has to be considered. The decay law follows from the differential equation

$$\frac{dN_2(t)}{dt} = N_1(t) \cdot \lambda_1 - N_2(t) \cdot \lambda_2 \quad (6)$$

With  $N_2(0) = 0$  follows

$$N_2(t) = N_1(0) \frac{\lambda_1}{\lambda_2 - \lambda_1} (e^{-\lambda_1 t} - e^{-\lambda_2 t}) \quad (7)$$

## 2.7 Statistics

For former calculations and estimation of uncertainties after the measurement, statistics like the Poisson distribution or the gaussian error propagation are needed and discribed in the following.

### Poisson Distribution

Measuring the half life of  $^{146}\text{Sm}$  leads to a small activity of a few counts per day. Because of this small number of decays statistics can be described by a poisson distribution [2]. Therefor the standard deviation is

$$\sigma_M = \sqrt{\bar{N}} \quad (8)$$

with the mean value for the number of decays  $\bar{N}$ . The relative error can be calculated as followed:

$$\frac{\sigma_M}{\bar{N}} = \frac{\sqrt{\bar{N}}}{\bar{N}} = \frac{1}{\sqrt{\bar{N}}} \quad (9)$$

### Gaussian Error Propagation

For further calculations, uncertainties have to be respected. These uncertainties are calculated applying the gaussian error propagation, because no covariances are expected. The error of a parameter  $x(a,b,c)$  is defined as

$$\Delta x = \sqrt{\left(\frac{\partial x}{\partial a} \Delta a\right)^2 + \left(\frac{\partial x}{\partial b} \Delta b\right)^2 + \left(\frac{\partial x}{\partial c} \Delta c\right)^2} \quad (10)$$



### 3 Calculations

Using the fundamental equations from Section 2 all parameters needed for activation and half life measurements can be determined. These parameters range from those needed for activation to the ones needed for the decay of  $^{146}\text{Pm}$  and  $^{146}\text{Sm}$ .

#### 3.1 Amount of Nd

The amount of particles can be calculated from

$$N = \frac{m}{M} N_A \cdot \text{abundance} \quad (11)$$

with the molar mass  $M$ , the overall mass  $m$  and the Avogadro constant  $N_A$ .

Having  $100\text{mg}$  of  $\text{Nd}_2\text{O}_3$  with an abundance of  $97.2\%$   $^{146}\text{Nd}$  and a molar mass of  $336.45\frac{\text{g}}{\text{mol}}$ , a number of  $N = 1.79 \cdot 10^{20}$   $\text{Nd}_2\text{O}_3$  particles results. To get the amount of Nd particles, this number has to be multiplied by 2. In total  $3.58 \cdot 10^{20}$  Nd particles can be activated.

#### 3.2 Activity for Significant Differentiation

Regarding to the poisson distribution enough decays have to be measured for a reliable value of the half life. To get an uncertainty below  $10\%$  for a measuring time of 40 days, an activity of at least 3 decays per day has to be chosen, which is obtained by using formula 9:

$$\sigma_M = \frac{1}{\sqrt{3\frac{\text{decays}}{d} \cdot 40d}} = 9.1\% \quad (12)$$

#### 3.3 Activity

The decay law for the reaction  $^{146}\text{Sm} \rightarrow ^{142}\text{Nd} + \alpha$  has to consider the formation and decay of  $^{146}\text{Sm}$ . Therefor a differential equation can be set up:

$$\frac{dN_{Sm}(t)}{dt} = N_{Pm}(t) \cdot \lambda_{Pm} - N_{Sm}(t) \cdot \lambda_{Sm} \quad (13)$$

With  $N_{Sm}(0) = 0$  it is

$$N_{Sm}(t) = N_{Pm}(0) \frac{\lambda_{Pm}}{\lambda_{Sm} - \lambda_{Pm}} (e^{-\lambda_{Pm}t} - e^{-\lambda_{Sm}t}) \quad (14)$$

Using the relation  $A = \lambda N$

$$A_{Sm}(t) = A_{Pm}(0) \frac{\lambda_{Sm}}{\lambda_{Sm} - \lambda_{Pm}} (e^{-\lambda_{Pm}t} - e^{-\lambda_{Sm}t}) \quad (15)$$

is achieved.

Eventually the start activity of  $^{146}\text{Pm}$  is

$$A_{Pm}(0) = \frac{A_{Sm}(t) \cdot (\lambda_{Sm} - \lambda_{Pm})}{\lambda_{Sm}(e^{-\lambda_{Pm}t} - e^{-\lambda_{Sm}t})} \quad (16)$$

With  $\lambda_{Pm} \gg \lambda_{Sm}$  ( $\lambda_{Pm} \approx 0.125$  and  $\lambda_{Sm} \approx 7 \cdot 10^{-9}$ ),  $A_{Pm}(0)$  can be approximated as:

$$A_{Pm}(0) = \frac{-A_{Sm}(t) \cdot \lambda_{Pm}}{\lambda_{Sm} \cdot (e^{-\lambda_{Pm}t} - 1)} \quad (17)$$

For measuring the half-life of  $^{146}\text{Sm}$ , at least 3 decays per day at the time of measurement  $t_m$  are necessary, compare Chapter 3.2. So an activity of

$$A_{Sm}(t_M) = \frac{3}{d} = 3.47 \cdot 10^{-5} \text{ Bq} \quad (18)$$

is obtained.

### 3.4 Flux and Irradiation Time

For measuring the half life of  $^{146}\text{Sm}$  accurately there have to be at least 3 decays per day. To get this activity it is necessary to know the required irradiation time. Therefore the flux is calculated by

$$\varphi = \frac{\dot{N}}{A_{\perp}} = \frac{I}{QA_{\perp}} \quad (19)$$

This flux can be inserted into the activity formula for activation:

$$A = N_0 \sigma \varphi (1 - e^{-\lambda T_{irr}}) \quad (20)$$

After rearranging the previous equation, an equation for the irradiation time is obtained:

$$T_{irr} = -\frac{\ln(1 - \frac{A}{N_0 \sigma \varphi})}{\lambda} \quad (21)$$

### 3.5 Calculation of the Half Life

Once the measurements are done, the half life of  $^{146}\text{Sm}$  will be calculated. Figure 4 illustrates the used parameters and how they relate the different nuclides.

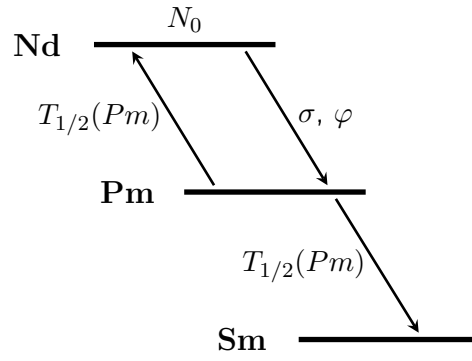


Figure 4: Scheme for used parameters from each step.

First, the activation has to be considered. Because of the long half life of  $^{146}\text{Pm}$  compared to the irradiation time, the exponential term in equation 20 can be neglected:

$$A_{Pm} = N_0 \sigma \varphi \quad (22)$$

and

$$N_{Pm} = A_{Pm} \cdot T_{irr} = N_0 \sigma \varphi T_{irr} \quad (23)$$

Using equation 6, neglecting the second term because of the small decay constant  $\lambda_{Sm} \approx 7 \cdot 10^{-9}$  and inserting equation 23

$$N_{Sm} = \frac{\ln 2}{T_{1/2}(Pm)} N_0 \sigma \varphi T_{irr} T_{wait} \quad (24)$$

is obtained.

With the number of  $^{146}\text{Sm}$ , the rate is given by:

$$R_{Sm} = N_{Sm} \frac{\ln 2}{T_{1/2}(Sm)} \quad (25)$$

To obtain the half life, equation 25 is transposed and equation 24 is inserted:

$$T_{1/2}(Sm) = \frac{\ln 2}{R_{Sm}} \frac{\ln 2}{T_{1/2}(Pm)} N_0 \sigma \varphi T_{irr} T_{wait} \quad (26)$$



## 4 Analysis

After theoretical consideration, suitable values for the parameter have to be found. Furthermore the activation problems will be discussed and the limit for the half life of the first excited state of  $^{146}\text{Nd}$  is estimated.

### 4.1 Choice of Activation Parameters

Regarding the high half life of  $^{146}\text{Sm}$  the amount of produced  $^{146}\text{Sm}$  should be maximized to gain a significant activity. Therefore the amount of  $^{146}\text{Pm}$  produced by proton activation of  $^{146}\text{Nd}$  enriched  $\text{Nd}_2\text{O}_3$  has to be sufficient.

#### Amount of $^{146}\text{Nd}$

The provided amount of  $\text{Nd}_2\text{O}_3$  is 100 mg, which corresponds to a number of  $3.58 \cdot 10^{20}$   $^{146}\text{Nd}$  particles. This mass is used for calculations, but there is no restriction to it. If the calculation show that the number of decays would not be sufficient otherwise, more  $\text{Nd}_2\text{O}_3$  can be acquired.

#### Proton Energy

To gain a sufficient amount of  $^{146}\text{Pm}$  a high cross section for the (p,n)-reaction has to be chosen. Due to the energy dependency of the cross section, an energy value with a high cross section has to be chosen.

Figure 5 shows the energy-dependency for the reaction  $^{146}\text{Nd}(p,x)^{146}\text{Pm}$ . The blue line is the summarized cross section for all reaction channels and the red line indicates the wanted cross section for  $^{146}\text{Nd}(p,n)^{146}\text{Pm}$ .

Up to energies of about 13 MeV, the summarized cross section is only composed of the cross section of the (p,n)-reaction. Therefore the cross sections given by Lebeda et al. [15] can be used.

For the cross section of  $^{146}\text{Nd}(p,n)^{146}\text{Pm}$  there is a maximum at 9 MeV, which is chosen for further calculations.

#### Beam Current

Choosing a high current leads to a high flux, which is direct proportional to the activity. But the higher the beam current is, the higher is the deposit of thermal power, [16]. Therefore a compromise between high activity and low current should be made, which is analyzed in Section 4.4.

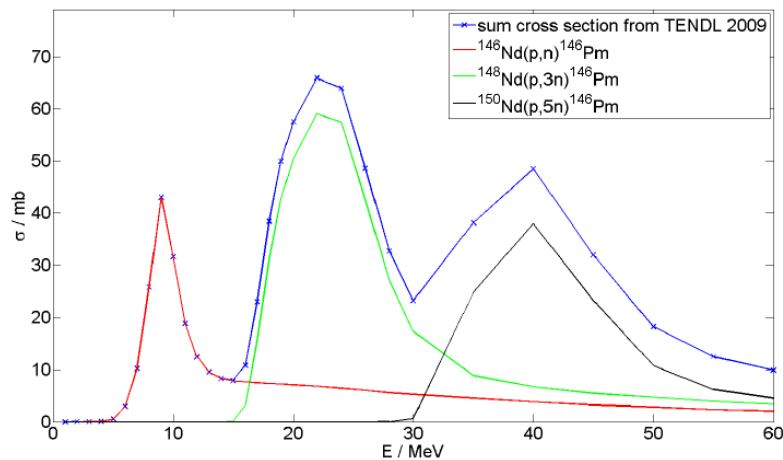


Figure 5: Cross-Section for the production of  $^{146}\text{Pm}$  via proton activation of  $^{\text{nat}}\text{Nd}$ , source [16]

## 4.2 Analysis of Activation Problems

To know, which reactions are possible, starting with  $^{146}\text{Nd}$ , the Q-value of this reaction has to be compared to the proton energy. Q-values for possible reactions are taken from [11].

With a proton energy of about 9 MeV, there are only two reactions left. On the one hand, there is the wanted reaction  $^{146}\text{Nd}(p, n)^{146}\text{Pm}$ . On the other hand, there can be the reaction  $^{146}\text{Nd}(p, \alpha n)^{142}\text{Pr}$  or  $^{142}\text{Pr}^{\text{m}}$ .

Regarding other isotopes of Nd in the sample, the Q-values of possible reactions [11] have to be reviewed. Table 2 shows all possible reactions for a proton energy of 9 MeV.

Some of these nuclides can be neglected due to a too low cross section, such as  $^{147}\text{Nd}$ ,  $^{149}\text{Nd}$ ,  $^{138}\text{Pr}^{\text{m}}$ ,  $^{139}\text{Pr}$ ,  $^{142}\text{Pr}$ ,  $^{147}\text{Pr}$  [15]. To get rid of nuclides with a short half life they will be especially stored for a few weeks, because after five half lives only 3.1 % of their activity is left and the resulting nuclides are stable. Therefore only  $^{143}\text{Pm}$ ,  $^{144}\text{Pm}$ ,  $^{148}\text{Pm}$  and  $^{148}\text{Pm}^{\text{m}}$  are left.

Other impurities can be identified via gamma spectroscopy if they are radionuclides. Therefore a gamma spectrum of the sample was recorded for 6 days at the Felsenkeller laboratory in Dresden. Figure 6 shows the analyzed spectrum, for which peaks were fitted and identified. Comparing the identified nuclides with the background usually gained with germanium detectors [3, 12], it can be seen that there are only decays from these environment nuclides.

Radionuclide	$T_{1/2}$	Decay mode	Nuclide	$T_{1/2}$ & Decay mode	Nuclide
$^{143}\text{Pm}$	265 d	EC	$^{143}\text{Nd}$	stable	
$^{144}\text{Pm}$	363 d	EC	$^{144}\text{Nd}$	$2.3 \cdot 10^{13}$ a, $\alpha$	$^{140}\text{Ce}$
$^{148}\text{Pm}$	5.368 d	$\beta^-$	$^{148}\text{Sm}$	$8 \cdot 10^{15}$ a, $\alpha$	$^{144}\text{Nd}$
$^{148}\text{Pm}^m$	41.3 d	$\beta^-$ , IT	$^{148}\text{Sm}$	$8 \cdot 10^{15}$ a, $\alpha$	$^{144}\text{Nd}$
$^{149}\text{Pm}$	53 h	$\beta^-$	$^{149}\text{Sm}$	stable	
$^{150}\text{Pm}$	1.68 h	$\beta^-$	$^{150}\text{Sm}$	stable	
$^{141}\text{Nd}$	2.49 h	EC	$^{141}\text{Pr}$	stable	
$^{141}\text{Nd}^m$	62 s	EC, IT	$^{141}\text{Pr}$	stable	
$^{147}\text{Nd}$	11 d	$\beta^-$	$^{147}\text{Pm}$	2.6 a, $\beta^-$	$^{147}\text{Sm}$
$^{149}\text{Nd}$	1.73 h	$\beta^-$	$^{149}\text{Pm}$	53.08 h, $\beta^-$	$^{149}\text{Sm}$
$^{138}\text{Pr}^m$	2.1 h	EC	$^{138}\text{Ce}$	stable	
$^{139}\text{Pr}$	4.4 h	EC	$^{139}\text{Ce}$	139, 7 d, EC	$^{139}\text{La}$
$^{140}\text{Pr}$	3.39 min	EC	$^{140}\text{Ce}$	stable	
$^{142}\text{Pr}$	19.1 h	$\beta^-$	$^{142}\text{Nd}$	stable	
		EC	$^{142}\text{Ce}$	$> 5 \cdot 10^{16}$ a, $2\beta$	$^{142}\text{Nd}$
$^{142}\text{Pr}^m$	14.6 min	IT	$^{142}\text{Pr}$		
$^{147}\text{Pr}$	13.4 min	$\beta^-$	$^{147}\text{Nd}$		

Table 2: Possible produced nuclides and their decays with 9 MeV protons.

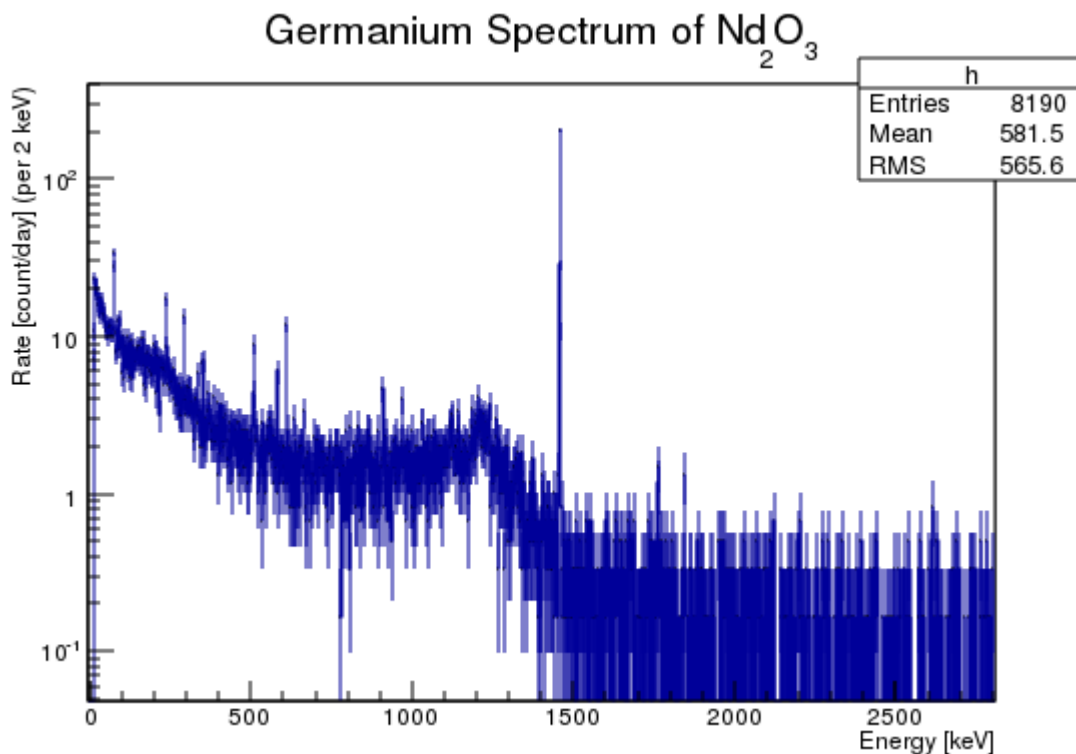


Figure 6: Gamma spectrum obtained from measurement in the Felsenkeller laboratory.

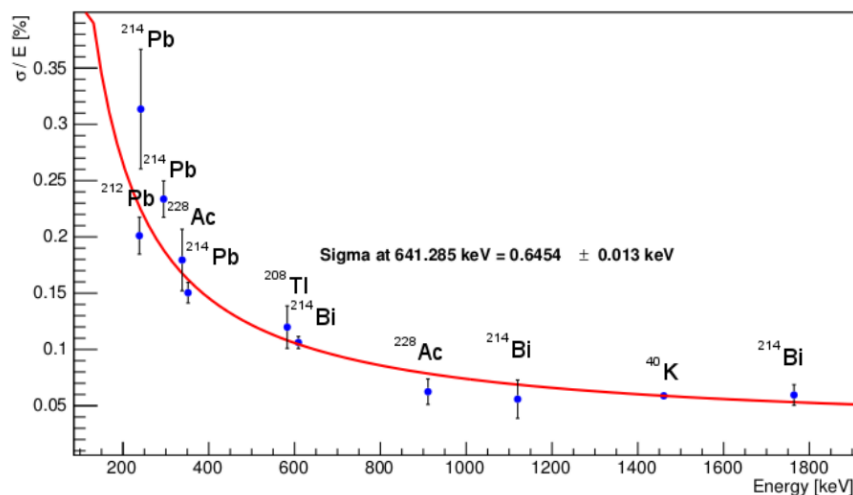


Figure 7: Energy resolution of the fitted peaks.

### 4.3 Half Life of the First Excited State of $^{146}\text{Nd}$

Within the scope of evaluating the gamma spectrum, the half life of the first excited state of  $^{146}\text{Nd}$  at 641.28 keV was determined. The decay of the first excited state of  $^{146}\text{Nd}$  has not been observed yet and decays via  $\alpha$ -decay to  $^{142}\text{Ce}$ .

The peaks seen in Figure 6 were fitted and using their statistics, a distribution of the standard deviation  $\sigma$  and their energy dependency can be set up and the energy resolution is obtained. It can be described by a function of the type

$$\frac{\sigma}{E} = \frac{a}{E} + b \quad (27)$$

illustrated in Figure 7.

Using the energy resolution function, the  $\sigma$  at 641.28 keV can be determined as  $0.06454 \pm 0.013$  keV. With the determined  $\sigma$  the region of interest (RoI) is set to

$$E_0 \pm 3\sigma(E_0) = 641.28\text{keV} \pm 3 \cdot 0.06454\text{keV} \quad (28)$$

To determine the background events in the RoI, the background in the region from 550 keV to 750 keV has been taken excluding the RoI as well as the 582 keV and 608 keV peaks. Afterwards the mean value was fitted as a straight line in the background and the average number of events per bin has been obtained. Figure 8 shows the distribution of background events, which follows a gaussian distribution and has a mean value of 9.883 events per bin.

For the RoI a background of around 39.5 events and a total amount of 49 events are determined. To get more accurate values for the half life of the first excited state of  $^{146}\text{Nd}$ , a Monte Carlo simulation of the  $^{40}\text{K}$  background was used. Therefore the  $^{40}\text{K}$



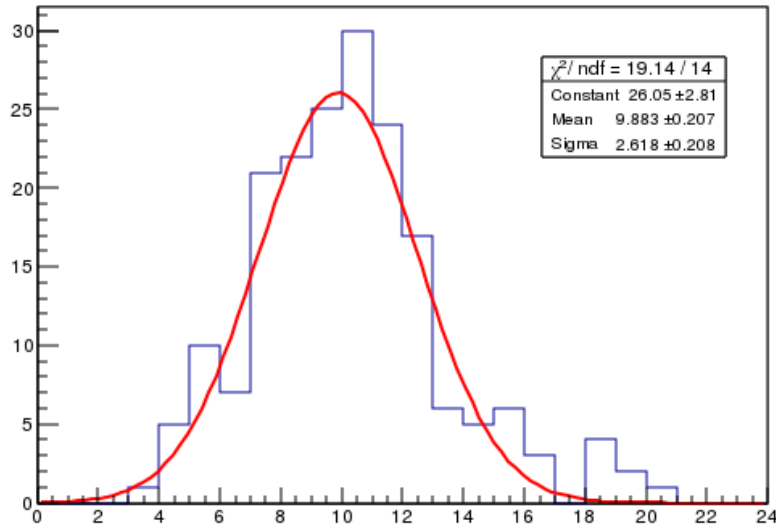


Figure 8: Distribution of the background events

events in the region of interest were subtracted from either the background and the total amount of events.

Using the Feldmann Cousins [6] class in ROOT, limits for the number of events for the  $\alpha$ -decay were calculated and converted into half life, considering the efficiency of the detector of 0.096. In this way a half life limit is calculated to  $1 \cdot 10^{18}$  years, using this simple counting method.

#### 4.4 Results from Programs

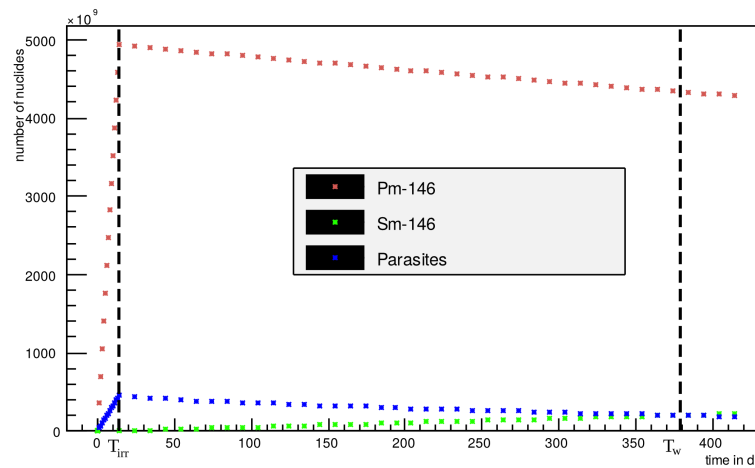
To estimate the activation time, a program was written with variation in current, waiting time and wanted activity of  $^{146}\text{Sm}$ . With these values, the needed activity of  $^{146}\text{Pm}$  and the time to activate the 100 mg  $\text{Nd}_2\text{O}_3$  are calculated. Table 3 shows calculated activation times for different input values. For the cross section a value of 35 mb is set, for the hit area 1  $\text{cm}^2$ .

Current/nA	$A_{Sm}(T_{measure})/\text{d}$	$T_{wait}/\text{a}$	$A_{Pm}(T_{act})/\text{Bq}$	$T_{act}/\text{d}$
50	3	0.5	10233.9	22.8
50	3	1	5276.79	11.7
50	3	1.5	3626.58	8.1
50	5	1.5	6044.3	13.5
100	3	0.5	10233.9	11.4
100	5	1.5	6044.3	6.7
100	10	1.5	12088.6	13.1

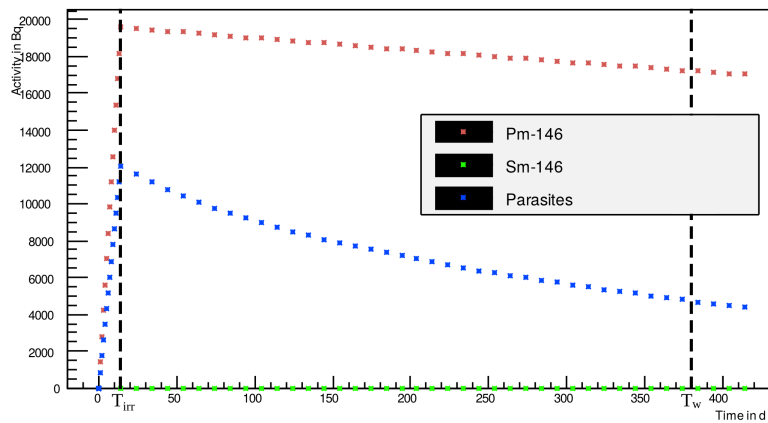
Table 3: Calculated values for different starting values.

Due to this work another program was written that shows the temporal development of the amount of nuclides produced by proton activation and their activity, which can be seen in Figure 9.

Looking at the number of parasitic nuclides, it is less than 10 % of the total number of produced nuclides. Due to the smaller half life of the parasitics compared to  $^{146}\text{Pm}$ , their activity is up to 38 % of the total activity, but decreases faster than the activity of  $^{146}\text{Pm}$ .



(a) Amount of nuclides as a function of time



(b) Activity as a function of time

Figure 9: Temporal development of  $^{146}\text{Pm}$ ,  $^{146}\text{Pm}$  and parasitic nuclides

The parasitic isotopes decay via electron capture and  $\beta^-$ -decay and the produced isotopes are long living  $\alpha$ -emitters. Due to the low number of produced  $\alpha$ -emitters and a half life several magnitudes larger than the half life of  $^{146}\text{Sm}$ , only few parasitic events should appear during the half life measurement.

## 5 Summary and Outlook

Calculations and estimations have been done for the whole process from activation to half life measurements. Within these, a proton energy of 9 MeV was chosen, because it goes along with a maximum for the cross section.

With a 9 MeV proton energy, 4 undesired nuclides can result from impurities, which are about 38 % of the whole activity directly after activation. The amount of these parasites is under 10 % of the total, but the activity is higher because of the lower half life compared to  $^{146}\text{Pm}$ . Because of this lower half life, the activity decays faster than for  $^{146}\text{Pm}$ .

An analysis of a gamma spectrum was performed to examine the  $\text{Nd}_2\text{O}_3$ -sample in respect to other impurities, where no radioactive impurities had been found. Using this gamma spectrum a limit for the half life of the first excited state of  $^{146}\text{Nd}$  has been calculated. The limit is  $1 \cdot 10^{18}$  years.

In the following, the sample will be prepared for activation. After that it will be activated and the half life will be measured using the alpha chamber, presented in Chapter 2.3.



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## **Erklärung**

Hiermit erkläre ich, dass ich diese Arbeit im Rahmen der Betreuung am Institut für Kern- und Teilchenphysik ohne unzulässige Hilfe Dritter verfasst und alle Quellen als solche gekennzeichnet habe.

Christiane Stengl  
Dresden, März 2015