Glowing green glass: Determining the uranium concentration in vaseline glass with a GM counter

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How much uranium is actually left in uranium glass after all these years since production? In this experiment, the current concentration of these radioactive isotopes is determined by using a Geiger-Müller counter, which can detect β-decay. The counts were measured over two iterations, each lasting a duration of 20 minutes, using the Geiger-Müller counter's built-in timer to measure this time span. The activity of the sample could then be determined using this data. The β-yield and geometric yield had to be taken into account to accurately arrive at this value, as well as the background radiation. Here it can be seen that the found value $C = (1.16 \pm 0.01) \cdot 10^{-2}\%$ is in conflict with the theoretical value of 0.5%-2.0%. The used method was validated through the use of a Poisson distribution, which confirmed that the setup was indeed detecting ionising radiation. The main cause of the deviation is the inability of taking the self-absorption into account due to the lack of comparable specimens with varying thicknesses being present. The use of such samples, along with a more suited GM counter is recommended in follow-up research in order to get a more accurate picture of the uranium concentration in the vaseline glass.

I. INTRODUCTION

Glowing green glass. No, it is not a vessel for some witch's concoction, but rather a common sight in the glass cabinets of the victorian upper class[1]. This vaseline glass, more commonly known as uranium glass gets its green glow from the presence of Uranium-oxide. In 1789 Martin Heinrich Klaproth discovered uranium, naming it after the then newly discovered planet Uranus[2]. Back then uranium was mainly used to stain glass a beautiful light green colour. This uranium glass was first produced in the 1830s, for which Czech industrialist Josef Reidel typically gets credited. The glass he invented is still being produced, though its popularity peaked in the 1880s.

Its radioactive properties were not discovered until 1996, by the scientist Henri Becquerel [3]. It was not until the mid 20th century that uranium was used as a fuel source for nuclear power, which to this day is still its primary function [2].

Since this type of glass has been around for a while, the question arises what the concentration is of that uranium in the glass. Uranium glass has been researched in the past, though there isn't much literature on determining its uranium content [4] [5]. This research paper focuses on answering the following question: What is the current concentration of uranium present in the vaseline glass?

A Geiger-Müller counter enables us to determine the uranium concentration in the glass by counting the decays over a certain time interval (see Theory). The expected result is a concentration of approximately between 0.5 and 2% by mass, which is the accepted theoretical value (see Theory).

II. THEORY

To determine the concentration of uranium in a vaseline glass sample at the hand of the counts given by a Geiger-Müller counter, one must make use of the following decay chains:

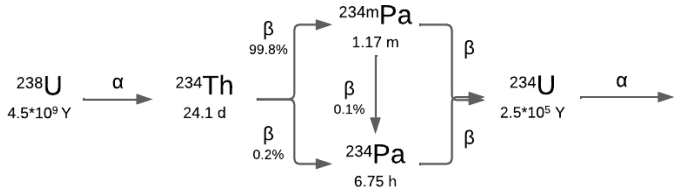

FIG. 1. The decay chain of uranium-238.

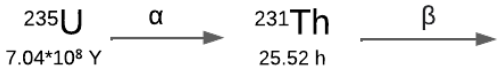

FIG. 2. The decay chain of uranium-235.

Due to the long half-lives of the further progeny in relation to those of the preceding elements in their respective decay chains, further steps of the decay chains can be considered not present. Furthermore, it is important to note that the Geiger-Müller counter is only able to detect β-particles and as such can only be used to determine the activity of the elements in the decay-chain that emit β-particles; Th-234, Pa-234m and Th-231 respectively. In this particular case the half-lives of the daughter radionuclides are orders of magnitude smaller than the those of the parent nuclides, which quickly leads to a secular equilibrium (i.e. the daughter nuclides are being produced at the same rate that they decay) [4]. As such, the activity of the uranium present in the glass can be determined at the hand of the measured

activity of its progeny. Typically, the concentration of uranium found in vaseline glass is between $0.5 - 2\%$ by weight [6].

With the known half lives for both isotopes ($t_{238} = (4.468 \pm 0.006) \cdot 10^9$ years, $t_{235} = (7.04 \pm 0.01) \cdot 10^8$ years [7]) the following formulas can be used:
The detected decay per second (R_c), given by the Geiger-Müller counter:

$$R_c = \frac{N_{count}}{\Delta t} \tag{1}$$

From this one can calculate a value for the total activity of the sample. This total activity can then be described as follows:

$$A(t) = A_0 \cdot 2^{-\frac{t}{t_{\frac{1}{2}}}} \tag{2}$$

With $A_0 = \frac{ln(2)}{t_{\frac{1}{2}}} \cdot N_0$ being the activity at $t = 0$ s and N_0 the amount of particles present at this time.
Furthermore, the corresponding amount of uranium present is:

$$N(t) = N_0 \cdot 2^{-\frac{t}{t_{\frac{1}{2}}}} \tag{3}$$

Since $m_n \approx m_p$, the current mass of the uranium can be calculated as follows:

$$m = A \cdot m_p \cdot N(t) \tag{4}$$

With A being the mass number, which differs per isotope.

It also should be noted that the glass itself will absorb some of the β-particles emitted by the present radionuclides. This self-absorption will vary substantially between different vaseline glass samples depending on the composition of the glass, and is thus very difficult to account for accurately.

III. METHOD

A. Experimental Setup

A set-up consisting of a Geiger-Müller counter and a uranium glass sample will be used in order to determine the uranium concentration. The sample used here has the shape of a lid for ease of measurement (see Figure 3).

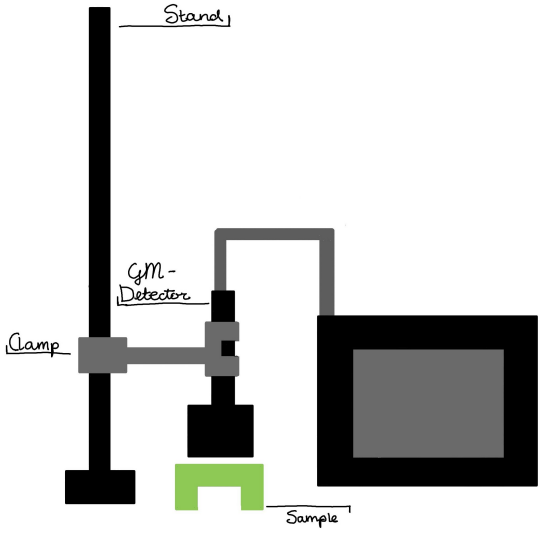

FIG. 3. The set-up, using a GM counter that only detects β-particles with a distance of at most one millimeter between the radioactive source and the detector. Here the generalised form of the sample is shown from a cross-section.

Here the varying value is the detected beta-decay from the GM counter. This counter is a Radhound SS315 probe [8]. The used efficiencies are those listed for the isotopes C-14, Pm-147 and Sr-90/Y-90 since $E_{d,Th-231} \approx E_{d,C-14}$, $E_{d,Th-234} \approx E_{d,Pm-147}$ and $E_{d,Pa-234m} \approx E_{d,YSr-90/Y-90}$ respectively [9]. Here, E_d is the decay energy per emitted β-particle from the mother nuclides. The average counts per second from the Geiger-Müller counter is displayed per count, therefore the uncertainty is one count.
It is important to note that not all beta-particles generated by the decay will be detected, as the paths they take are unpredictable and random. To correct for this, the geometric yield (G) and β-yield (ϵ) should be taken into account [10]. G is the ratio of the amount of electrons having reached the detector to the actual amount emitted by the source (see Appendix). ϵ is the ratio of the amount of detected beta particles and the amount beta particles that have reached the detector (see Appendix). The corresponding uncertainty is estimated to be ten percent of the found value of ϵ given the amount of assumptions that have had to be made in order to arrive at these values.
A ruler is needed to determine the dimensions of the detector and the used sample. The corresponding uncertainty $u(x)$ is 0.05 cm per measurement made.
A weighing scale is used to determine the mass of the sample. The value measured by this weighing scale will have an uncertainty of $u(m) = 0.02$ g.

B. Procedure

An accurate approximation of the background radiation can be made by measuring the counts for 5 minutes

without any radioactive material present below the detector. After having determined the background radiation, the radiation of the sample will be measured. The closest value to the actual activity will be found by measuring the counts of the sample two times, each iteration lasting 20 minutes. Here, the built-in timer will be used to ensure the smallest deviation from the set-point time intervals. The distance between the detector and source will remain unchanged by holding the detector with a clamp attached to a stand. Taking all these factors into account ensures that the data acquired from these measurements will be accurate.

C. Analysis

For accurate measurements, the background radiation (N_b) has to be taken into account. This turns equation 1 into:

$$R_c = \frac{N_{count}}{\Delta t} - \frac{N_b}{\Delta t_b} \tag{5}$$

Given the duration of the determined time intervals, previous equations 2 and 3 become:

$$N(t) = \frac{A(t) \cdot t_{\frac{1}{2}}}{ln(2)} \tag{6}$$

Taking the geometric yield and β-yield into account leads to the following equation:

$$A(t) = \frac{R_c}{\epsilon \cdot G} \tag{7}$$

With the given ratios, the activity per corresponding isotope becomes:

$$A_{238}(t) = \frac{1}{2} \cdot 0.96A(t) \tag{8}$$

$$A_{235}(t) = 0.04A(t) \tag{9}$$

This gives:

$$N_{238}(t) = \frac{A_{238}(t) \cdot t_{238}}{ln(2)} \tag{10}$$

$$N_{235}(t) = \frac{A_{235}(t) \cdot t_{235}}{ln(2)} \tag{11}$$

Using equations 4, 10 and 11, the masses per isotope can be determined and we arrive at the following equation for the current concentration (C) of the total uranium in the glass:

$$C = \frac{m_{238} + m_{235}}{m_{sample}} \tag{12}$$

Here m_{238} and m_{235} are the corresponding masses per isotope present in the glass. m_{sample} is the total mass of the uranium glass sample used for measurements.

In order to confirm the validity of the measurements, a Poisson distribution will be fitted to a histogram of one of the measurements in Python [10]. Here the amount of counts per short time interval of ten seconds will be measured across 20 minutes. The uncertainties of the values calculated above, will all be determined using the calculus approach (see Appendix).

IV. RESULTS

After having executed the experiment as described in the method, two values of R_c were measured. These were 35666 and 35485 counts respectively, both counted over a time span of 20 minutes. At the time of the measurement, the background radiation came out at a value of 207 counts measured over 301 seconds. The count rate according to the Geiger-Müller counter, when taking the average of the two values and the background radiation into account, was 29 counts per second (cps). The calculated β-yield value is 0.12±0.02 and the geometric yield is 0.39±0.04. After having factored in those efficiencies of the measuring equipment, the value for the activity ($A(t)$) was found to be $646 \pm 1 \cdot 10^2$ Bq. To verify if radioactive decay was actually measured, a histogram was constructed, which was fitted to a Poisson distribution (see Figure 4). The histogram in the figure closely follows the poisson distribution, with a mean (λ) of 296 counts per 10 seconds. From this, the resulting activity can be used to determine the concentration of both isotopes (U-235 and U-238): $C = (1.16 \pm 0.01) \cdot 10^{-2}\%$.

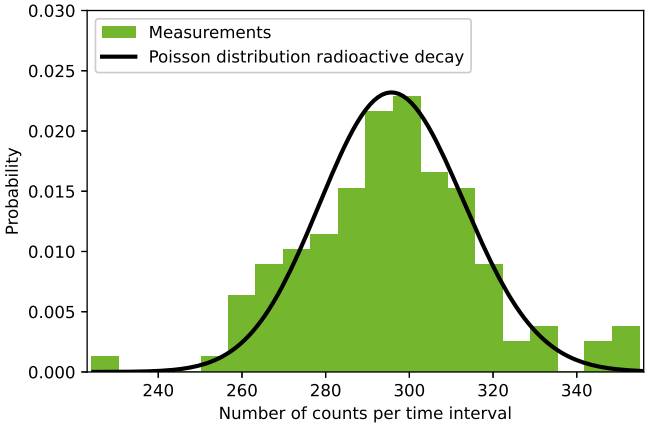

FIG. 4. Here the radioactive Decay of Th-234, Th-231 and Pa-234m are depicted, with each time interval lasting 10 seconds, with a total duration of 20 minutes. The histogram is fitted to a Poisson distribution, with a mean (λ) of 296 counts per 10 seconds.

V. DISCUSSION

The mean (λ) derived from the fitted Poisson distribution was 29.6 counts per second, which is close to the 29 counts per seconds shown by the GM-counter. It is noticeable that the probability is highest around the 290 counts per 10 seconds and gets lower the further one gets from the mean. After having compared the fitted measurements in a Poisson distribution with the found count rate of 29 counts per second, the method can be deemed correctly executed. This means that the setup did indeed detect ionising radiation coming from the vaseline glass sample.

The found concentration of uranium in vaseline glass, $C = (1.16 \pm 0.01) \cdot 10^{-2}\%$, conflicts with the expected value of 0.5%-2.0%.

The assumption of the general shape of the glass specimen can be one of the causes of this deviating value. As such, the determined value for the geometric yield (G) could be incorrect by some margin. Furthermore, the β-yield (ϵ) is based on comparing energy-levels of the given isotopes in the data-sheet with the ones present in the uranium glass. These two assumptions clarify the high uncertainty in the activity measured.

It was also not possible to account for the self-absorption of the β-decay within the sample itself, due to the lack of comparable samples with varying thicknesses. This would have likely led to a substantially larger calculated concentration.

Taking this self-absorption into account is recommended for follow-up research. This can be accomplished by having comparable samples available with the only difference being the thickness. With this sample then being of a more uniform (disc) shape, the calculated volume, and therefore also the geometric yield, will be more accurate. Furthermore, a better suited GM counter is recommended to avoid comparing energy levels E_d of the decay, and as such also the efficiencies of the probe, with different radionuclides.

VI. CONCLUSION

The current concentration of uranium isotopes within a vaseline glass sample was to be determined in this experiment. Since the original production of the glass, the concentration has not been properly measured. Having measured the weight of the sample, this value can be found by using a Geiger-Müller counter with a β-detector. When fitting a Poisson distribution to the measurements, it can be seen that the found count rate from the GM-counter (29 cps) is almost the same as the one shown in the graph (29.6 cps). This indicates that the setup did indeed measure ionising radiation. By using the known masses and half-lives of the isotopes, the found activity values are used to derive the concentration (C) within the glass specimen: $C = (1.16 \pm 0.01) \cdot 10^{-2}\%$. This is in conflict with the theoretical value of 0.5%-2.0%.

This deviation can be caused by the assumptions for the volume calculations and the best comparable efficiencies. Furthermore, the lack of comparable samples with different thicknesses meant the self absorption could not be accurately accounted for. By having such glass specimens with a more uniform (disc) shape and a better suited GM counter available, such shortcomings can be prevented in follow-up research.

APPENDIX

A. Calculating yields

Calculation for the β-yield:

$$\epsilon = 0.96 \cdot \frac{\rho_{C-14} + \rho_{Y-90/Sr-90}}{2} + 0.04 \cdot \rho_{Pm-147} \quad (13)$$

ρ is the efficiency from the data-sheet with the corresponding particle.

Calculation for the geometric yield:

$$G = \frac{R}{V} \quad (14)$$

Here, R is the amount of volume being measured by the detector and V the total volume of the sample.

B. Calculus approach

Below are the uncertainties calculated by the calculus approach:

$$u(G) = \sqrt{u(R)^2 + u(V)^2} \quad (15)$$

$$u(A_{238}) = \sqrt{(\frac{u(\epsilon)}{\epsilon})^2 + (\frac{u(G)}{G})^2} \cdot A_{238} \quad (16)$$

$$u(A_{235}) = \sqrt{(\frac{u(\epsilon)}{\epsilon})^2 + (\frac{u(G)}{G})^2} \cdot A_{235} \quad (17)$$

$$u(N_{238}) = \sqrt{(\frac{u(A_{238})}{A_{238}})^2 + (\frac{u(t_{238})}{t_{238}})^2} \cdot N_{238} \quad (18)$$

$$u(N_{235}) = \sqrt{(\frac{u(A_{235})}{A_{235}})^2 + (\frac{u(t_{235})}{t_{235}})^2} \cdot N_{235} \quad (19)$$

$$u(m_{238}) = \frac{u(N_{238})}{N_{238}} \cdot m_{238} \quad (20)$$

$$u(m_{235}) = \frac{u(N_{235})}{N_{235}} \cdot m_{235} \quad (21)$$

$$u(M) = \sqrt{u(m_{238})^2 + u(m_{235})^2} \quad (22)$$

$$u(C) = \sqrt{(\frac{u(M)}{M})^2 + (\frac{u(mass)}{mass})^2} \cdot C \quad (23)$$

$$(24)$$

Here, $mass$ and $u(mass)$ correspond with the total mass of the used sample.

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