Programmatic Thermal Neutron Flux Calibration for Neutron Generators

A Major Qualifying Project Submitted to the Faculty of Worcester Polytechnic Institute in partial fulfillment of the requirements for the Degree of Bachelor of Science in Physics by

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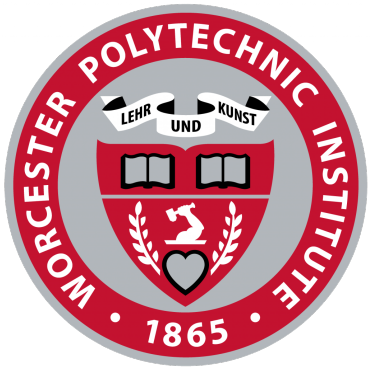

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Abstract

Neutron activation analysis (NAA) allows for precise and accurate measurements of isotope concentrations within a sample. The technique is versatile in its applications in nuclear research, materials science, etc. A major constraint in NAA is calibration time, which currently requires a full sample activation and gamma spectroscopy analysis. This project addresses calibration time by applying a programmatic activation function that allows for instantaneous flux measurement of the generator. The iterative method discussed in this report shows promise to reduce calibration time to a fraction of the current industry standard while maintaining high accuracy and precision.

Contents

Acknowledgments

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Executive Summary

Neutron activation is a method of transforming a stable isotope into a radioactive isotope. By bombarding the sample with free thermal neutrons, the nuclei of a sample can absorb these neutrons therefore changing the isotope. If radioactive, the newly formed isotope then decays according to a decay rate dependent on their half-life, $t_{1/2}$. Neutron activation is of interest to researchers for its ability to precisely and accurately determine the concentrations and activities of elements within a sample. This technique, known as neutron activation analysis (NAA) offers superior precision and accuracy to any other method reasonably available to researchers. With NAA's ease of use, high replicability, and versatility in measuring numerous isotope concentrations simultaneously, the technique was quickly adopted and is widely used in the fields of material science, criticality safety, and nuclear physics research.

Still, NAA has its limitations. After a sample has been activated, it will continue to be radioactive for a period of time. The use of NAA also requires the use of a neutron source and a gamma spectroscopy system, making the technique costly in terms of equipment. Further, this equipment must be calibrated on a regular basis to determine neutron flux and account for ambient activity. These barriers make NAA difficult to access and use regularly.

Calibration of equipment seems to be a glaring challenge in need of modernization. Since the first mention of neutron activation by G. Hevesy and Hilde Levi in 1936[1], the world has entered an age of computers. Over the past several decades, computational methods have been adapted and developed for data analysis, optimization, and calibration. Therefore we propose a new calibration method of neutron sources in this paper. Using MATLAB, a programmatic solution to the measurement of neutron flux emitted from WPI's neutron generator promises to cut calibration time to a small fraction of where it stands

today. The method is easily generalized and thus has potential to streamline the calibration of neutron generators.

1 Introduction and Background

1.1 Review of Radioactivity

Radioactivity or radioactive decay is defined as the process by which energetic particles are ejected by unstable atomic nuclei[2]. The rate of this decay is determined by the half-life $t_{1/2}$ of the isotope in question. Based on this, we are able to determine the number of decayed isotopes at any time based only on the starting mass and the half-life via equation 1. One question that immediately jumps to mind is the scenario involving a single radioactive isotope. Half-life only tells us the time it will take for half of the substance to decay, and a single particle cannot be in a state of "half decayed". In this case, the decay of the particle is stochastic; entirely random. The half-life will indicate how likely the particle is to decay in that time, but there is no way to predict exactly when decay will occur[3]. During the decay of an atomic nucleus, varying forms of radiation are emitted based on the element isotope (referred to as a "parent nuclide") as it transitions into a more stable daughter nuclide.

1.1.1 Alpha Decay

The most massive example of radiation is alpha decay. Among the first to be identified by Ernest Rutherford in 1903, alpha radiation takes place when the parent nuclide ejects two protons and two neutrons, effectively yielding a more stable daughter nuclide of smaller atomic mass and a He^{2+} nucleus. Generally, this decay takes the form

$$^{A}_{Z}X \rightarrow ^{A-4}_{Z-2}Y + ^{4}_{2}\alpha.$$

For an example of this, consider the decay of Thorium-228 to Radium-224:

$$^{228}_{90}Th \rightarrow ^{224}_{88} Ra +^{4}_{2} \alpha.$$

Alpha decay is non-ionizing, and is most common among heavy elements and almost exclusively occurs in elements heavier than tellurium (atomic number 104-109). However, one isotope of beryllium, beryllium-8, tends to decay into two alpha particles[4].

Due to the large mass of alpha particles, alpha radiation is more easily shielded than other forms of radiation. Alpha radiation typically does not penetrate skin or pose a health risk to biological subjects unless ingested, injected, or inhaled[5].

1.1.2 Beta Decay

Beta decay is a non-ionizing form of radiation characterized by the emission of an electron (beta-minus decay) or positron (beta-plus decay) from the nucleus. Like alpha decay, beta decay results in a change in element as a neutron becomes a proton or vice-versa. However, due to a minute difference in mass between the proton and the neutron, energy conservation appears to be violated. Pauli proposed the existence of the neutrino in 1930 to account for this discrepancy and has since been experimentally verified[3]. As such, beta decay takes the general form:

$$^{A}_{Z}X \rightarrow ^{A}_{Z-1} Y + \beta^{+} + \nu_{e},$$

or, in the case of beat-minus decay:

$$^{A}_{Z}X \rightarrow ^{A}_{Z+1} Y + \beta^{-} + \bar{\nu_{e}},$$

where β^+ is a positron, β^- is an electron, ν_e is an electron neutrino, and $\bar{\nu_e}$ is an electron antineutrino.

While low-energy β particles cannot penetrate the skin and are thus largely harmless unless ingested, high-energy β particles can penetrate several centimeters of tissue before being stopped. β particle emissions can thus cause severe damage and burning to the tissue, particularly if ingested [5].

1.1.3 Gamma Decay

We arrive now at a subject of more immediate relevance to our methodology. Gamma decay involves the emission of a high-energy photon from the nucleus. It most commonly takes place during alpha or beta decay. Gamma radiation is fundamentally distinguished from alpha and beta decay in that gamma decay is ionizing and does not change the isotope or element of the source. For example, consider the decay of Cobalt-60 to Nickel-60:

$$^{60}_{27}Co \rightarrow ^{60m}_{28} Ni + e^- + \bar{\nu} + \gamma + 1.17MeV.$$

Here, the change of the cobalt nucleus to a nickel nucleus is due to the emission of a β^- particle, and the change caused a release of energy in the form of a gamma-frequency photon of energy 1.17 MeV. But we now have a very excited nickel-60 that would like to decay further toward the ground state. This follows the process:

$$^{60m}_{28}Ni \rightarrow ^{60}_{28} Ni + \gamma + 1.33MeV,$$

where the suffix m in the mass number denotes an excited state of the nucleus.

It is immediately noticeable that the energies of photons emitted from the nucleus are significantly higher than even those emitted by inner-shell electrons

during orbital transitions, hence their classification as gamma energy photons. This discrepancy is due to the enormous electromagnetic repulsion experienced by proximal protons in the nucleus, particularly as they shift toward a more "comfortable" arrangement[1]. This brings to intuitive light that gamma decay often follows isomeric transitions, as the nucleus of the newly formed element is rarely in such a geometry that it does not favor a transition to a lower energy state. In most cases, there are significantly more excited states than stable states of an atomic nucleus[6].

Gamma emissions from the nucleus typically occur on the order of 10^{-15} seconds. An excited nucleus that does not emit a gamma-range photon in the femtosecond range is considered to be in a metastable state, where it can stay in this excited state for widely varying periods of time. In most cases this time remains well below one second, but an interesting example is found in tantalum-180m, which can remain in metastability for upwards of 10^{15} years[6]!

Several dangers of gamma emissions arise from its penetrating strength and high energy. It cannot be effectively shielded by protective suits and presents a great danger to biological matter in high doses[7].

Gamma radiation has been leveraged for use in research. The utility of gamma emissions stretches from the fields of cosmology[8] to densitometry and woodworking[9]. One of the more extraordinary applications of gamma emissions is found in gamma spectroscopy, which has allowed for precise and accurate measurements of gamma emitters in a source.

[1]This would be an excellent opportunity to discuss the strong nuclear force and its raw strength such that it is able to overcome such ridiculous electromagnetic force and allow for the existence of the vast number of elements we observe in the universe, but that is beyond the scope of this research.

1.1.4 Neutron Radiation

Neutron radiation is an ionizing radiation that involves the ejection of free neutrons from the nucleus. This ejection is most commonly a product of nuclear fusion or fission, but can occur in nuclei with an excess of neutrons following beta decay[10]. After emission, free neutrons have a mean lifetime of 887.7 seconds, after which they decay into a proton, electron, and anti-electron neutrino[11].

Free neutrons are able to be absorbed by atomic nuclei, altering the isotope and inducing radioactivity in the subject. The radioactive samples decay according to the equation:

$$N_t = N_0 e^{\lambda t} \tag{1}$$

where N_t is the quantity of radioactive isotopes at time t, N_0 is the initial quantity of radioactive isotopes, and λ is the decay constant defined by

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

The most effective shields against neutron radiation are light nuclei, which mitigate the momentum of fast neutrons. Materials such as water, concrete, and hydrogen-rich polymers make excellent shields against free neutrons[11].

While neutron radiation can be highly destructive to biological matter[12], the method of neutron activation analysis leverages this induced radioactivity through neutron absorption in tandem with gamma spectroscopy to efficiently analyze the compositions of activated sources.

1.2 Neutron Activation Analysis

Neutron activation analysis (NAA) refers to the process by which samples are irradiated using a neutron source and then analyzed by their decay products.

The activity of the sample is determined by the equation:

$$A = N\phi\sigma(1 - e^{-\lambda t}), \tag{3}$$

where A is the activity of the sample in bacquerels, N is the number of sample atoms, ϕ is neutron flux density in neutrons per square centimeters per second, σ is the nuclear cross section in square centimeters, λ is the decay constant in decays per second, and t is time in seconds.

NAA allows for detection of trace samples of $> 1mg$ with accuracy in the realm of 2% and precision of the similar magnitude[13]. In the decades since the discovery of neutron activation, the unique decay signatures of hundreds of isotopes have been researched for use in NAA (see appendix A for table of frequently used radioisotopes)[14]. The neutron source used for NAA in this particular research is a deuterium-deuterium generator. Through the ionization and collision of deuterium atoms, a yield of helium and free neutrons are achieved in the following reaction:

$$^2H +^2 H \rightarrow^3 He + N.$$

The particular model of neutron generator used by WPI is Adelphi's DD110M generator, which is capable of achieving a flux on the order of $10^6 - 10^7$ Neutrons per square centimeter[15]. To achieve an accurate number for flux, a calibration must be done using a sample with a well-known cross-section and decay spectrum. Gold foil is a popular sample material, and is used for calibration of WPI's neutron generator.

1.2.1 Adelphi DD110M Generator

Adelphi's DD110M Neutron Generator, shown in figure 1, is used by WPI for research in neutron imaging. WPI also hosts external research groups that use the generator for their own research. According to the specifications offered by the manufacturer, the DD110M is capable of producing both thermal ($\phi = 10^6 - 10^7 N/cm^2/s$) and fast ($\phi = 10^8 - 10^9 N/cm^2/s$ neutrons, which rivals much larger nuclear reactors while remaining intrinsically safer than fission reactors due to its use of deuterium-deuterium collisions for neutron production. Interestingly, while the thermal neutron output of the DD110M is listed to be $10^7 N/cm^2/s$, the researchers at WPI have been able to achieve thermal neutron flux of $10^8 N/cm^2/s$ by creating fast neutrons and thermalizing them after production via an aluminum wall and concrete slabs to slow fast neutrons down to the thermal range[16].

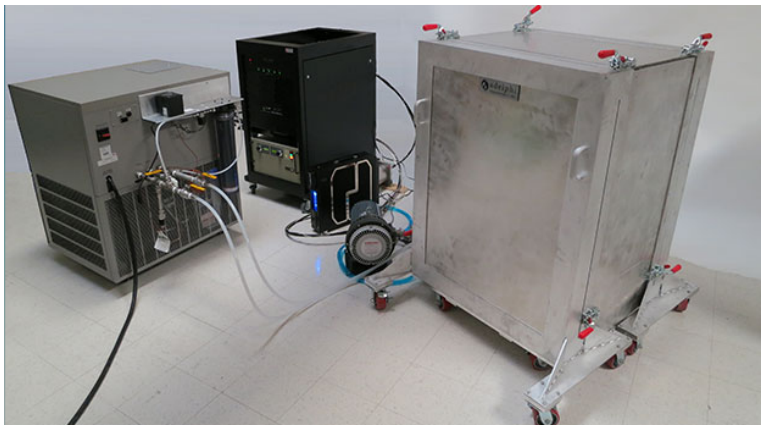

Figure 1: DD110M Neutron Generator manufactured by Adelphi Technologies for use in WPI's radiation laboratory.

In order to calibrate the DD110M, and more generally all neutron generators, an activation analysis must be performed on a well known material. After 600

seconds of irradiation of the sample, it is analyzed using a gamma spectrometer (shown in figure 2) to determine the number of activated nuclides, and average flux in the region of the sample is determined by the equation:

$$\overline{\phi} = (C\lambda)/((N\sigma\epsilon Y(1 - e^{(-\lambda t_a)}(1 - e^{(-\lambda t_c)})e^{(-\lambda t_d)})), \tag{4}$$

where C is the gamma counts measured during analysis, N is the number of sample atoms, Y is the empirically determined yield, t_a is the time the sample has been irradiated, t_c is the time the sample spends in the gamma spectrometer, and t_d is the time it takes to move the sample from the generator target area to the gamma spectrometer for analysis[16]. This calibration must be performed regularly, particularly if the generator is operating at variable voltages. With this in mind, it is apparent that this calibration method becomes tedious, as the absolute shortest time this calibration can take is 30 minutes when including sample preparation and storage time. The goal of this project is found in reducing the amount of time this calibration takes by implementing an iterative function derived from the experimentally determined relationship between neutron flux and electric current[16].

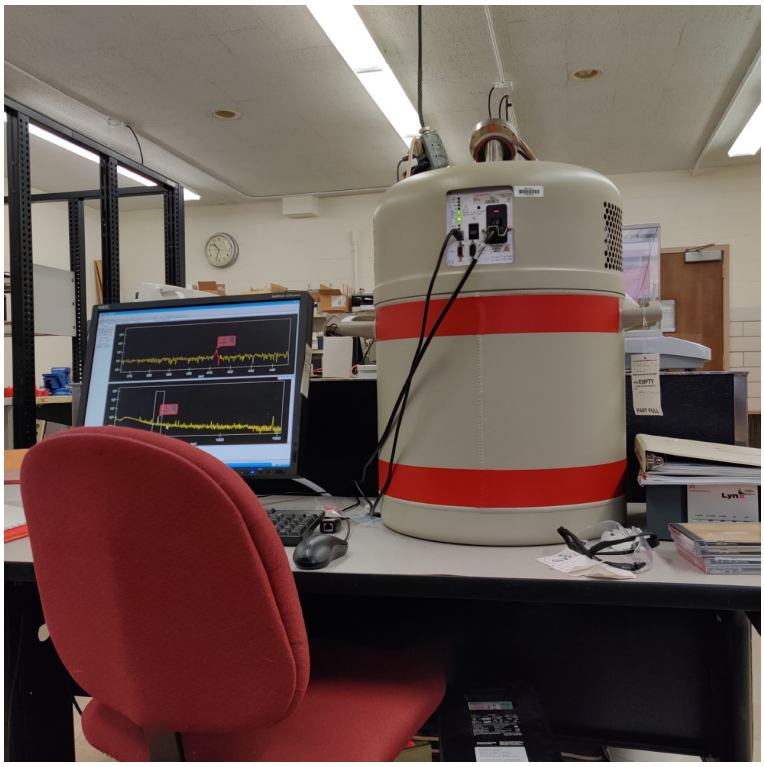

Figure 2: Broad energy germanium detector (BEGe) Gamma Spectrometer manufactured by Mirion Technologies for use in WPI's radiation laboratory.

2 Methodology

2.1 Building a MATLAB Script for Average Neutron Flux

The first step toward building an accurate iterative function to determine flux was to generate a script that calculates the average flux using the known method of equation 4 (see appendix A). This method is accurate when considering an irradiated sample with a long half life relative to the analysis time for calibration. The time between sample irradiation and gamma detection is negligible and can be approximated to 600 seconds. All other parameters such as N, ϕ, σ, λ, etc. are not approximated and are precise values given to the function as user input.

The average flux thus offers an accurate empirical baseline for comparison to the iterative function.

Empirical flux measurements were gathered via gold foil activation. A 0.05 gram sheet of gold foil is massed before being irradiated by the neutron generator for 600 seconds. During this time, an approximately continuous flux is assumed. However, due to irregular arcing and railing of the electric current, during which flux is known to be zero, an uncertainty is introduced on the order of $< 1\%$. This uncertainty is small enough to be negligible due to the long half life of gold and exponential activity function. Thus, our approximation of average neutron flux across the irradiation period is accurate given a stable electric current.

Included in the script is an iterable that counts the total number of arcs and rails the generator undergoes during a sample activation. This information is of interest to the lab as a whole for purposes of analyzing generator behavior.

2.2 Correlating Neutron Flux to Neutron Beam Current

It is known that there exists a positively correlated relationship between the operating current and neutron fluency[16]. Using this knowledge, a series of gold foil activations were done in order to characterize this relationship. These measurements were taken in late 2017, however, and generator functionality has changed significantly since then. This is taken into consideration in the error analysis.

This relationship is of critical importance to the programming of an iterative activation function. It allows for the removal of gold foil activation as a necessary step in determining mean flux and a real time calculation of flux at any time t as a function of operating current.

2.3 Obtaining an Iterative Activation Function

The iterative activation function was derived from the relationship

$$\frac{dn}{dt} = N\sigma\overline{\phi} - \lambda n, \tag{5}$$

where $\frac{dn}{dt}$ is the number of nuclides dn activated in time dt. From here, we can discretize dt as some Δt and we have

$$\Delta n = [N\sigma\overline{\phi} - \lambda n]\Delta t.$$

We multiply both sides by λ and take ϕ as some function of current $\phi(I(t))$

$$\Delta n\lambda = [N\lambda\sigma\phi(I(t)) - \lambda^2 n]\Delta t,$$

but

$$\lambda n = A$$

and thus

$$\lambda\Delta n = \Delta A$$

hence our discretized iterative function:

$$A_{i+1} = [N\lambda\sigma m(I(t_i) - I_0) - \lambda A_i]\Delta t, \tag{6}$$

where I_0 represents the minimum current necessary to produce flux.

2.4 Validation of Activation Function

After the programmatic activation function has been developed, it can be experimentally verified using gold foil activation. After performing a gold foil activation, the generator data can be analyzed via our iterative function and compared to the results given by gamma spectroscopy.

The efficiency of the programmatic method is undeniable, but its accuracy and efficacy must be within acceptable error for the method to hold validity.

2.5 Error Analysis

Due to scholarly discrepancies in the uncertainties of the cross-section, decay constant, and gamma detection processes, it is difficult to reasonably approximate the error associated with NAA without straying beyond the scope of this research. According to a comprehensive study published in 2019 by the Annals of Nuclear Energy [17], their is a systemic error in NAA measurements of 7%.

Further, there were several instances of operational hiccups during data acquisition. Through the summer and fall of 2019, there were parts being changed out of WPI's neutron generator which caused a change in the target operating current of the generator. This had invalidated previously collected data necessary to calculate the slope factor for the iterative function, and data had to be rerecorded.

Other sources of error that are difficult to quantify include stochastic neutron scattering and absorption by the collimator, and Adelphi DD110M software bugs that cause the generator data to become unreadable during arc events. The iterative function attempts to address this issue by checking for arc events in each iteration and properly adjusting flux according to the last known operational status of the generator.

For the purposes of this research as a simple proof of concept, error will be

taken as the minimum acceptable value of 7%.

3 Results

3.1 Calculation of Slope Factor

Three measurements were taken at current values of 7 mA, 8 mA, and 10 mA. The dependence of flux upon operating current was found to be linear with a slope factor $m \simeq 7.9498 * 10^6 cm^{-2} * s^{-1} * mA^{-1}$. This relationship is modeled in figure 3. The x-intercept, when solved for, shows I_0 to theoretically rest at $5.8570mA$. However, the smallest current value at which flux has been experimentally observed was found at $5.5mA$. This indicates a need for more trials to collect data points.

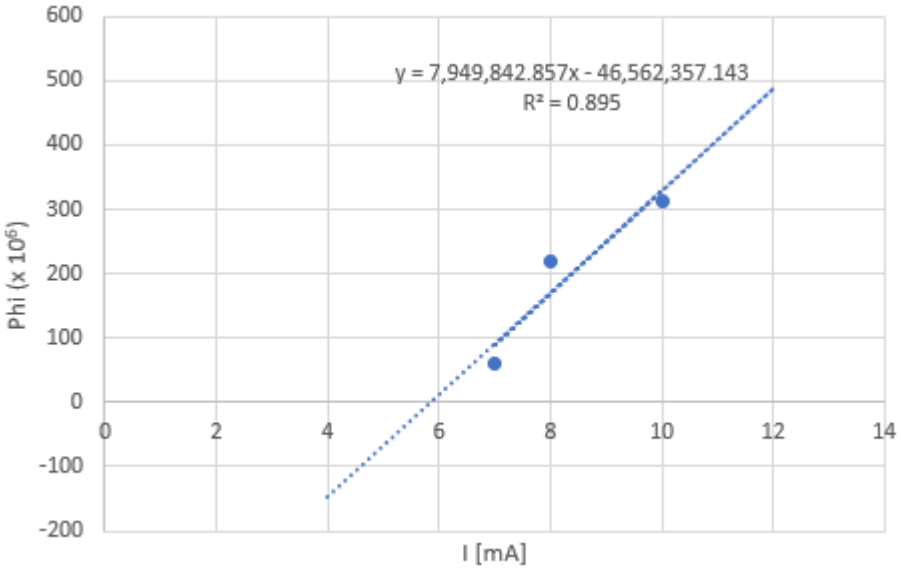

Figure 3: Neutron flux as a function of current during gold foil activation.

3.2 Gold Foil Activation

Flux determined via GFA is shown in table 1. Due to the generator's scarce availability, only three trials were able to be included.

Table 1: Flux Calculated via Gold Foil Activation.

Trial	Mass (g)	Target $I(mA)$	$t_i(hr)$	$t_d(hr)$	$t_c(hr)$	C_{Au}	$\overline{\phi}(n * cm^2 s^{-1})$
1	0.0498	10.00	0.16667	0.025	0.16667	515	$3.13600 * 10^7$
2	0.0485	8.00	0.16667	0.05	0.16667	348	$2.17646 * 10^7$
3	0.0745	7.00	0.41667	0.3333	0.5000	1077	$4.89300 * 10^6$

3.3 Iterative Activation Function

Flux calculated via the iterative activation function is shown in table 2.

Table 2: Flux Calculated via Iterative Function

Trial	Mass (g)	Target $I(mA)$	$A_f(n * s^{-1})$	$\overline{\phi}(n * cm^2 s^{-1})$	Error
1	0.0498	10.00	832.729	$3.102 * 10^7$	1.084%
2	0.0485	8.00	619.259	$2.368 * 10^7$	8.824%
3	0.0745	7.00	241.410	$4.858 * 10^6$	0.715%

4 Conclusions

By deriving an iterative method of determining neutron flux from operating current, an accurate and efficient supplement to NAA has been demonstrated. While there remains work to be done in optimizing the method for laboratory use, preliminary results show that accuracy within the realm of $< 1\%$ is achievable. Due to the Covid-19 pandemic, more data was not able to be collected at the time of submission. A wider range of target currents should be tested to achieve a more dependable standard deviation.

5 Discussion

Despite restricted sample size, the iterative method shows promise as a way to significantly reduce calibration time for the DD110M neutron generator. Due to the method's simplicity, the relationship between neutron fluency and operating current can be easily generalized for application in any nuclear physics lab that currently uses NAA for calibration. While calibration via NAA will no longer be regularly necessary, NAA should be used whenever operating conditions change, such as the target's distance from the source, individual components of the generator, etc.

Another merit to be found in the iterative method is the ability to calculate the instantaneous flux at any second of the generator's operation. This allows for improved monitoring of neutron fluency and measurement of localized radioactivity surrounding the generator without dependence on a neutron detector.

Work can be further done to optimize this solution for ease of use. This script has the potential to run in real time without manual input of generator log data.

Appendix A: Source Code for GFA Flux Calculation

```
count=input('enter count');
Y=.95; %What is actual yield?
ti=input('enter ti');
tc=input('enter tc');
td=input('enter td');
epsilon=1.065e-3;
sigma=9.87e-23;
N=input('enter number of sample atoms');
lambda=2.98e-6; %Decay constant
Data=NGData.Variables;
ta=0; %seconds spent irradiated
Ta=0; %seconds spent decaying
taCounter=0; %counter for ta
TaCounter=0; %counter for Ta
Switches=0; %Number of times sample alternates between activation and decay
Arcs=0; %Number of times the generator arcs, starting decay in the sample
for j=2:1:size(Data,1)
    if Data(j,5)==0 && Data(j-1,5)~=0% || Data(j,3)==0 && Data(j-1,3)~=0
        Switches=Switches+1;
        Arcs=Arcs+1;
    elseif Data(j,5)~=0 && Data(j-1,5)==0
        Switches=Switches+1;
    end
end


flux_avg=(count*lambda)/((N*sigma*epsilon*Y*(1-exp(-lambda*ti))*exp(-lambda*td)*(1-exp(-lambda*tc))));
```

Appendix B: Source Code for Iterative Activation Function

```
N=input('enter number of sample atoms');
ti=input('enter ti');
Data=NGData.Variables;
lambda=2.98e-6;
sigma=9.87e-23;
m=7.949e6; %Slope of phi vs. I
Ai=0; %Activation=0 at t=0
flux_array=zeros(1,size(Data,1)); %store flux at each iteration
AiArray=zeros(1,size(Data,1)); %Store activation at each iteration
for q=1:size(Data,1)
    if Ai>=0 && Data(q,5)<38 && Data(q,5)>5.5
        Ai=Ai+(N*sigma*lambda*(m*(Data(q,5)-5.5)))-(lambda*Ai);
        AiArray(q)=Ai;
        flux_i=(Ai-AiArray(q-1))/(N*sigma*(1-exp(-lambda)));
        flux_array(q)=flux_i;
    elseif Ai>=0 && Data(q,5)==0 && Data(q+1,5)>5.5 && Data(q-1,5)>5.5
        Ai=Ai+(N*sigma*lambda*(m*(Data(q-1,5)-5.5)))-(lambda*Ai);
        AiArray(q)=Ai;
        flux_i=(Ai-AiArray(q-1))/(N*sigma*(1-exp(-lambda)));
        flux_array(q)=flux_i;
    else
        if Ai>0
            Ai=Ai-(lambda*Ai);
            AiArray(q)=Ai;
            flux_i=0;
            flux_array(q)=flux_i;
        else
            Ai=0;
            AiArray(q)=Ai;
```

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