Experiment #4 - Automation of Nuclear Decay Rate Measurements

Introduction

Model. The decay of a population of radionuclides is found to obey a differential equation of the following form:

$$-\frac{dN}{dt} = kN \tag{1}$$

Where N is the number of nuclides present as a function of time, and k is the decay rate in inverse time and can be evaluated from the half-time of the first-order reaction (*i.e.*, the time for the population of nuclides to decay to one-half their original value):

$$k = \frac{\ln_e 2}{\tau_{1/2}} = \frac{0.693}{\tau_{1/2}} \tag{2}$$

When the first equation is integrated from time t = 0 to t, subject to the initial condition, $N(0) = N_o$, the following equation results:

$$N(t) = N_0 e^{-kt} \tag{3}$$

The population $N(t)$ is not measured directly. Instead, the "activity" is determined by measuring the number of nuclear disintigrations per unit of time with the aid of radiation detector such as a Geiger counter. Activity measurements, $A(t)$, are proportional to the time rate of change of the population. Thus,

$$A(t) = -\frac{dN(t)}{dt} = kN_o e^{-kt} = A_o e^{-kt} \tag{4}$$

where A_o is the initial activity of the sample at time t =0. Often, there is some background radiation arising from radioactive sources that do not change with time. This results in the addition of a constant term in the above equation:

$$A(t) = A_o e^{-kt} + B \tag{5}$$

where the constant B accounts for the background activity.

Measurement Uncertainty. Unfortunately, as in all experimental measurements, there is some irreproducibility in the data. In the case of nuclear decay curves, this irreproducibility arises solely from the random nature of the decay process, which leads to a type of uncertainty known as Poisson or counting noise. There are many ways to justify the mathematical form of Poisson noise, but the following considerations provide

an intuitively satisfying approach and lend themselves to a simulation of nuclear decay curves that includes noise.

In the above equations, we have described the macroscopic behavior of an ensemble (collection) of atoms. What happens at the level of one atom and what does the decay curve mean for a single atom? We postulate that for a single atom, the decay curve actually represents the probability that the atom, newly formed at time $t = 0$, will suddenly convert to a new species at a later time, $t = t_d$. Clearly, from the form of an exponential decay curve, t_d can take on any value between zero and infinity, with $t = 0$, the most probable value. From a single decay time measurement on a single atom, we can tell nothing about the decay curve. However, if we repeat the measurement many, many times, we can form a histogram of values of t_d that begins to resemble the decay curve.

Simulation. How can we simulate the decay of single nucleus? The answer is to use a random number generator that returns a value from a set of numbers that obey the exponential distribution of the nuclear decay law. Unfortunately, in LabVIEW, we only have a random number generator whose values are uniformly distributed along the interval 0 to 1. However, we can easily convert this uniform random distribution to an exponential distribution by means of the transformation $-(1/k)*ln(rand)$, where rand is a specific value taken from the set of uniformly distributed random numbers.

LabVIEW can be readily employed to simulate a nuclear decay curve, including noise. Essentially, we use a do loop to generate a series of numbers, say 1000, from a random number generator which is exponentially weighted, by using the transformation $y = (1/k)*ln(rand)$ inside a formula node. Once the 1000 numbers are generated, we can form a histogram with histogram.vi. and then plot the results. When this histogram is plotted, the character of the noise becomes apparent, as shown in the front panel of the vi Decay_Sim.vi.
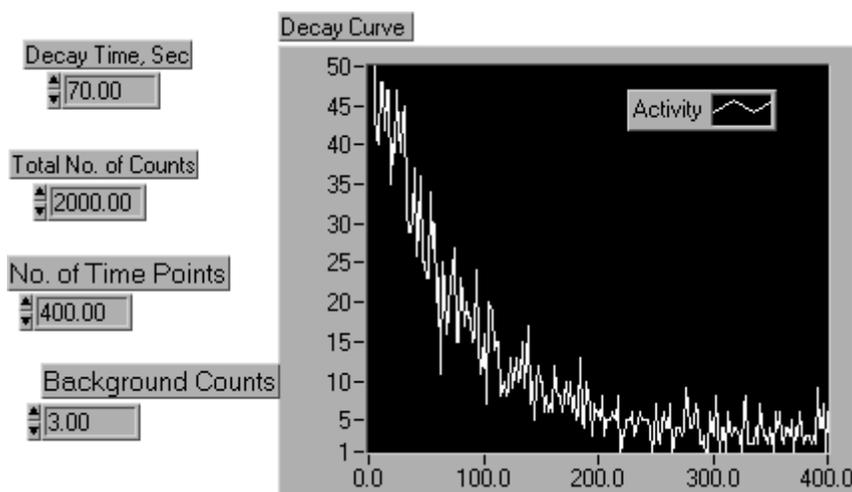
Figure 1A Simulated Nuclear Decay Curve, Activity in Decompositions/sec vs. Time, sec
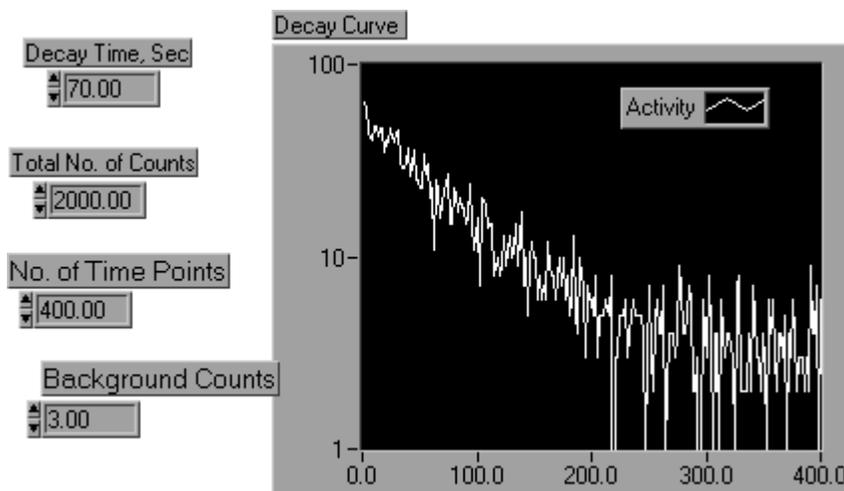
Figure 1B Simulated Nuclear Decay Curve, Activity in Log (Decompositions/sec) vs. Time, sec

Why are these curves so noisy? The answer is that we have counted only a small number of events. As described above, this type of uncertainty is called the counting error or Poisson noise. It arises from the uncertainty in the time a nucleus will take to release a radioactive particle. One of the important features of Poisson noise is that the standard deviation in the count is equal to the square root of the count.

How can we simulate the background counts and their contribution to the noise? The answer is to generate a separate histogram for the background using the same number of points (bins) in the histogram. The total number of random numbers to generate is just the background counts per unit interval times the number of bins in the histogram.

Chemical System. In this experiment you will measure the half-life of the nuclide ^{234}Pa (protactinium). This specie is part of the decay chain of natural ^{238}U which is summarized below:

$$^{238}_{92}U \xrightarrow[\alpha(4.18)]{4.49 \times 10^9\,yr} {}^{234}_{90}Th \xrightarrow[\beta^-(0.193)]{24.1\,da} {}^{234}_{91}Pa \xrightarrow[\beta^-(2.31)]{1.18\,min}$$

$$^{234}_{91}Pa \xrightarrow[\beta^-(0.16,0.55)]{6.7hr} {}^{234}_{92}U \xrightarrow[\alpha(4.76)]{2.48 \times 10^5\,yr} {}^{230}_{90}Th \xrightarrow[\alpha(4.69)]{8.0 \times 10^4\,yr}$$

$$(5\alpha \text{ and } 4\beta \text{ decays}) -\!-> {}^{208}_{82}Pb$$
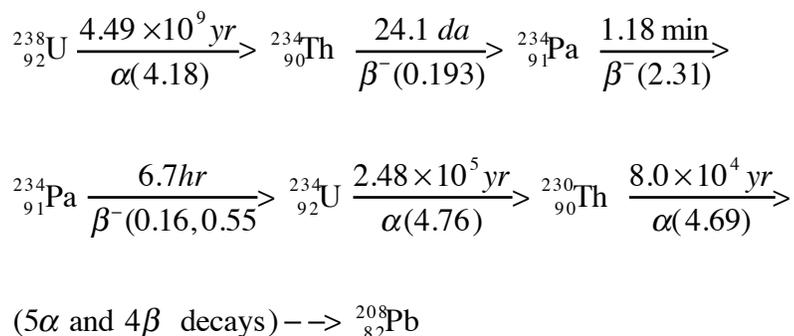
Of the various activities early in the decay chain which are in equilibrium with $^{238}_{92}U$, the 1.18 min $^{234}_{91}Pa$ has the most energetic beta radiations, and can thus be readily detected in

the presence of other activities. To optimize the detection of this nuclide, as well as to provide an isolated sample of nuclides suitable for a half-life determination, advantage is taken of the fact that protactinium can be extracted from 7 *N* aqueous HCl into methyl isobutyl ketone under conditions which leave the uranium and thorium mainly in the (heavier) aqueous layer.

Approximately one gram of $^{238}_{92}U$ is dissolved in 15 ml of 7 N HCl and placed in a 50-ml polyethylene bottle. To this is added 15 ml of hexone, and the assembly is tightly closed. Lead shielding is wrapped around the tube so that the Geiger counter sees only the organic layer as shown in Figure 4 below. This amount yields a concentration of the $^{234}_{91}Pa$ high enough to obtain a good signal to noise ratio.

To measure the decay curve, the polyethylene bottle is withdrawn from the counting station, shaken vigorously for about 30 sec, and returned to the lead shield at the same time at which a laboratory timer is started. After allowing exactly 30 sec to elapse for the two phases to separate, counts are taken at 1 sec intervals for 400 sec. The data are then analyzed by least squares fitting of the decay curve to the model of Equation 1.

The three parameters, A_0, k and B are varied in order to obtain the best fit of the model to the data. The offset term, B is due to background caused by the extraction of some long-lived activities other than ^{234}Pa into the aqueous layer.

Instrumentation and Computer Interfacing. The beta ray (electron) emitted when the Protactinium decays, is detected by a Geiger counter. This device emits a pulse of 10^8 - 10^{10} electrons per beta in a period of 20 microseconds. The pulse is emitted on a background of 750 volts. Electronics in the counter then convert the analog pulse into a digital logic pulse of 3.5-5.0 volts with duration of 50 microseconds. Such pulses are suitable for counting by the timer-counter facility that is part of your DAQ board.

The counter/timer facility consists of two independent 24 bit up/down counters. We will use one counter to measure the number of pulses that occur in a pre-determined time period. In practice, as shown in figure 2, the signal is connected to the source terminal of the counter and the counting is started and stopped by use of the gate terminal. When the gate signal is high counting occurs and when low, counting is suspended. The number of pulses detected is available on the out terminal. These signals you need for performing the experiment are available on the breakout box.
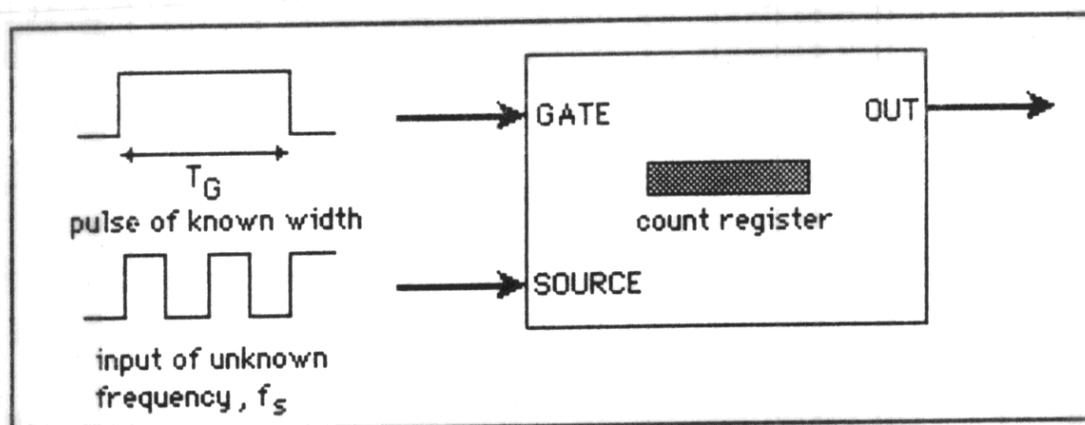
Figure 2. Measuring the number of pulses during a counting interval.

Software. You will perform the counting operation using the vi, "Measure Frequency". Timing will be generated by the crystal clock on the DAQ board using a separate counter (see Figure 3). You will be provided with a handout which describes this vi.
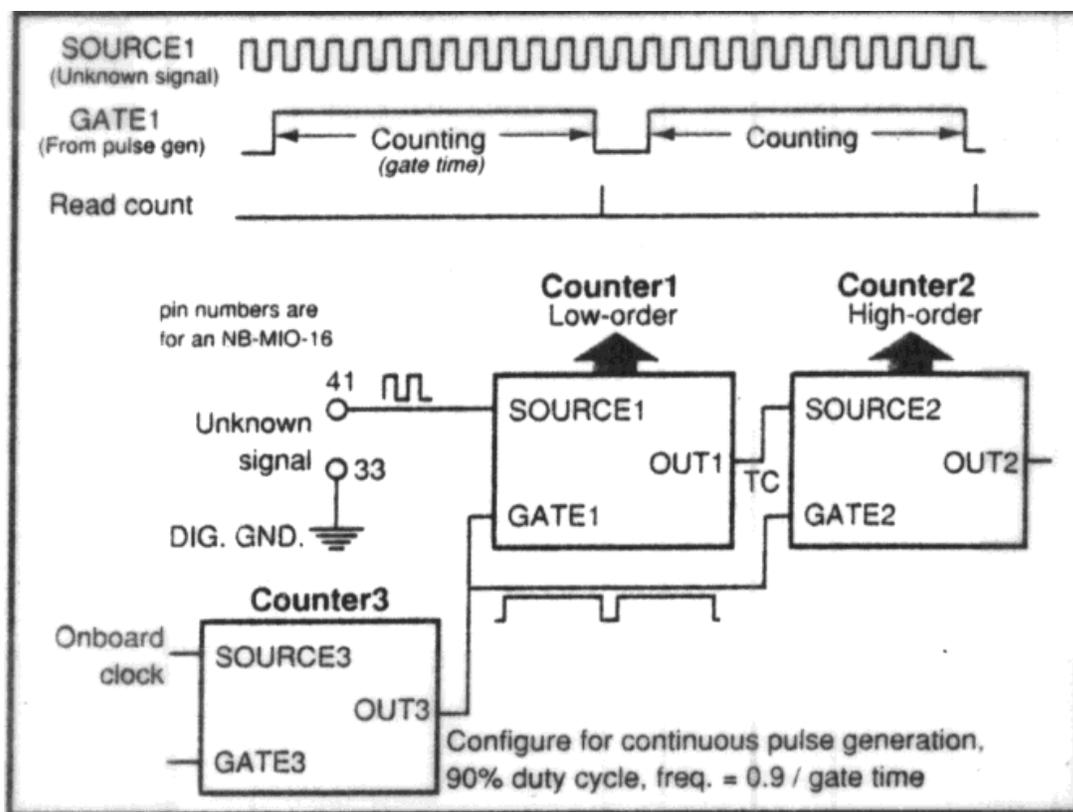
Figure 3. Counting with DAQ crystal clock timing.

Data Analysis. You will fit the model $A(t) = A_o e^{-kt} + B$ to your data by varying the parameters A_o, B and k, until the model is as close to the data as possible using the

criterion of least squares. Since this is a non-linear problem, you will use the vi "Levenberg Marquardt.vi". You will be given a handout which describes this vi. Your final plot should show the data as unconnected circles and the best fit theory as a solid line.

<div align="center">**Procedure**</div>

Part 1 - Simulate a Nuclear Decay Curve.

A simulation vi is available for down-loading from the class web site. It follows the theory outlined in the first part of the introduction. Your vi's front panel with data output in the form of a graph should look like Figures 1A and B. Run this vi under various conditions of input parameters to get a feel for how the appearance of the decay curve varies. Start from the conditions given in Figure 1. What happens to the curve as more counts are obtained? What happens if the background becomes greater? Evaluate the effect of varying the number of time points. Are all of these variations consistent with the fact that the standard deviation in the Poisson noise in each time interval is just the square root of the counts collected?

Part 2 - Development of an Automated Experiment to Acquire Decay Data and Analyze It.

The experimental set-up is given below and you can see it in lab at your instructor's station.
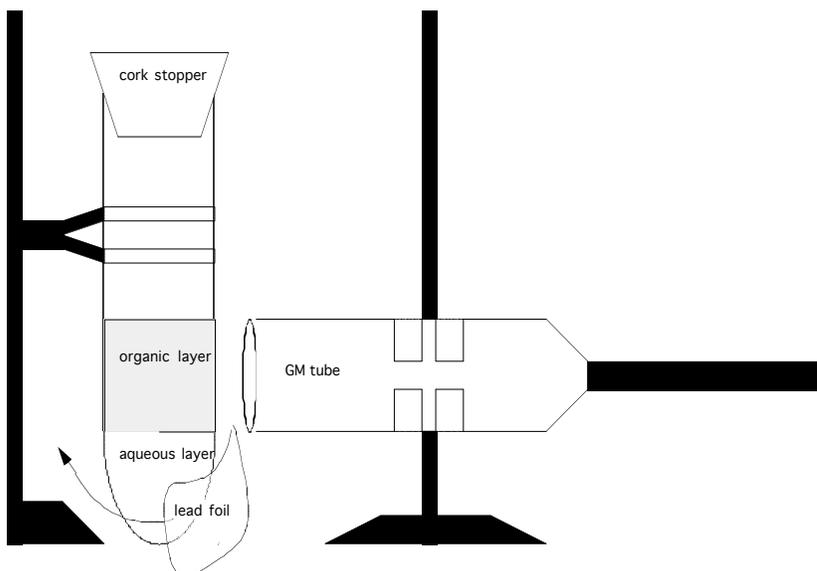
Figure 4. Experimental Setup Chem 464 Lab 4 - Timing and Counting

There is a Geiger counter at each station. Turn the Geiger counter on using the black knob in the center. Adjust the setting to the X1 position. Read the top scale of the meter:

its units are in CPM (counts per minute). The AUD switch allows you to hear each count made by the Geiger counter. Please perform the experiment with this switch on off.

Before you develop your vi, make sure that the counter works and the sample contains radioactive material. Notice that your radioactive sample is in a polyethylene bottle with a screw cap. Ensure that the cap is tightly fastened. To begin the experiment, you need to extract the active nuclide, $^{234}_{91}Pa$ into the organic layer. This is accomplished by shaking the bottle vigorously for 30 seconds.

As soon as you stop shaking, place the bottle in front of the Geiger tube as per Figure 4. However, do not collect data for 30 seconds; this delay will allow time for the phases to separate fully. Notice the interface between the aqueous and organic layers of the solution in the bottle. During the 30 second wait, note that there is a piece of lead foil which is wrapped around the bottom of the bottle to cover the aqueous layer. Adjust the Geiger tube so it is at the level of the organic layer, point it towards the organic layer and bring it in contact with the bottle.

During this 30 second preparation period verify the settings on the Geiger counter. Be sure that the F/S switch on the Geiger counter is on the F position. You may turn the AUD switch to On for a moment if you want to hear the Geiger counter responding to radiation. In general keep the Geiger counter silent in order to avoid disturbing others.

Read the CPM (counts per minute) scale on the meter every 20 seconds for 400 seconds. If the pointer goes off scale, increase the scale reading to X10 or X100 to bring the pointer back on scale. Be sure to multiply the reading you take from the meter by the appropriate factor before recording your values. During the series of measurements, you may notice the counts per minute falling to below one. In this case increase the sensitivity by changing the setting to 0.1 CPM. At this setting, you will probably need to set the F/S setting to S.

Just for fun, plot your data on a piece of graph paper. Better yet, use Excel to plot the data. Show the data to your instructor to make sure it is acceptable. Remember, the computer version of the experiment should obtain the same type of data.

You are now ready to hook up the counter to the data acquisition board via the breakout panel. The data is available to the outside world as TTL digital logic pulses on the BNC connector on the side of the Geiger counter. Connect this signal to your scope's channel 1 with a BNC "tee" connector and then use a cable with clip leads to connect to the breakout panel. Connect the black clip lead to pin 44, DGND and the red clip lead to pin 42, SOURCE 1. Also, to use the hardware clock for generating timing pulses, you will need to connect pin 2 on the breakout panel (GPCTR0_OUT (OUT 5)) to pin 41 on the breakout panel (GPCTR1_GATE (GATE 1)).

Now develop your vi for data acquisition. Base the it on the vi, "Measure Frequency." Set the number of data points and the data sampling interval from the front panel. Enclose the

Measure Frequency.vi inside a for loop set to execute a number of times equal to the number of data points. Use the data sampling parameter to set the gate interval for the counter. Inside the for loop, connect the output of the counter to a waveform chart so you can see how the data acquisition is progressing. At the end of the data acquisition provide a means to store the data as a spreadsheet file. Now you are ready to try out your vi.

Once the data acquisition vi is developed, you are ready to develop an analysis vi. You will first set up a means to read in the data using the Read from Spreadsheet File.vi. Next you will plot the input data an XY graph with the data points as open circles with no lines connecting them. Then you will enter the initial guesses for the parameters on the front panel. Then the Levenberg Marquardt.vi will take over for a maximum number of iterations you set from the front panel. You will report your results as (a) the optimized set of parameters on the front panel, and (b) a plot, which you put as a solid line on the pre-existing graph of the data. You should use a weighted least squares fit to get the best results. You may take a peek at the front panel of the instructor's vi.

For your report, hand in your two vis as both hard copy, using Print Documentation with typical data shown and on diskette. Your vis should be properly documented, including critical parts of the components of the diagram. For comparison, synthesize some artificial data with the same values of the parameters, A_o, B and k as you obtained from the least squares fitting of the data. Then submit your artificial data to the analysis program. Do you get the same results for the optimum values from the experiment? Repeat the simulation experiment a number of times to get a feel for how well the analysis is expected to do. Report your results in a table of values obtained for A_o, B and k for both simulated and real data. If you have time, run the experiment a number of times to get error estimates for the three parameters: A_o, B and k. Report your error estimate.

Before leaving, get a background count from your hands to make sure there is negligible residual radioactivity on your person. Contact your TA immediately if you measure an elevated count (>500 CPM).

Finally, be sure to turn off the Geiger counter.