

Investigating Nuclear Decay Phenomena

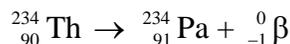
Background

Ordinary chemical reactions involve changes in the outer electron structure of atoms. The particles in the nucleus (protons and neutrons) are typically far removed from these changes. When atoms of sodium react with chlorine molecules the number of *nucleons* (nuclear particles) remains unchanged in each atom. While the sodium atoms become ions with positive charges, they are still sodium in some fundamental sense. Their identity is fixed by the number of protons in the nucleus. Different sodium atoms may contain different numbers of neutrons but these do not have the same "identity" characteristic as the number of protons.

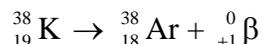
Atoms of the same element with different numbers of neutrons are called *isotopes* (from the Greek for "same place", meaning their position in the Periodic Table). The chemical properties of different isotopes of the same element are very similar. Sometimes the subtle differences can be used in experimental work to closely examine chemical behavior. For example, heavier isotopes of atoms involved in rate determining steps may slow down a reaction. This recognition can aid in the elucidation of a reaction mechanism.

Some isotopes have special characteristics that are not related to their chemical properties but rather to the stability of the nucleus itself. Observation of neutron-proton combinations in isotopes reveals that some ratios result in stable configurations, presumably related to an internal structure of the nucleus, while others result in unstable arrangements which revert to more stable forms over time. The latter group of nuclei are described as *radioactive*. In a radioactive nucleus, the weak electrostatic repulsive forces overcome the strong nuclear force and generally small pieces of the nucleus are ejected until the neutron-proton ratio is that of a stable configuration. In contrast to chemical processes, these *nuclear* reactions result in a change in the number and type of nucleons and thus the identity of the elements. The outer electrons are left virtually untouched.

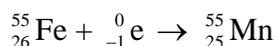
The variation in neutron-proton ratios suggests a variety of pathways by which a radioactive nucleus may become more stable. In nuclei which are "neutron-rich", these excess particles can be transformed into protons by the ejection of a particle (indistinguishable from an extra-nuclear electron) known as a *beta particle* (β^-):



"Proton-rich" nuclei may become more stable by transforming a proton into a neutron and ejecting the anti-matter equivalent of an electron, a *positron* (β^+):



An alternate pathway involves the capture of an extra-nuclear electron (usually from the 1s orbital), also enabling the conversion of a proton into a neutron:



Adapted from: [LPC Radiation Lab](http://lpc1.clpccd.cc.ca.us/lpc/physics/pdf/mitchrad.pdf), Mitch Moffet: <http://lpc1.clpccd.cc.ca.us/lpc/physics/pdf/mitchrad.pdf>,

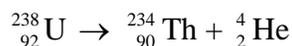
[Gamma Ray Spectroscopy](http://artemis.austincollege.edu/acad/physics/Irobin/gamma%20exp%2098.pdf), Modern Physics Lab Manual: [http://artemis.austincollege.edu/acad/physics/Irobin/gamma exp 98.pdf](http://artemis.austincollege.edu/acad/physics/Irobin/gamma%20exp%2098.pdf) ,

[Automation of Nuclear Decay Rate Measurements](http://courses.washington.edu/jbcallis/labs/C464_Lab4_LabMan_Sp-02.doc), J.B. Callis: http://courses.washington.edu/jbcallis/labs/C464_Lab4_LabMan_Sp-02.doc

[Statistics of Nuclear Decay](http://www.csupomona.edu/~pbsiegel/www/bio431/Chapter2.doc), Peter Siegel: <http://www.csupomona.edu/~pbsiegel/www/bio431/Chapter2.doc>

[Experiments in Nuclear Science](#), Chase, Rituper and Sulcoski

Nuclei that have mass numbers higher than 209 frequently shed excess mass in larger increments by emitting helium ions (consisting of two protons and two neutrons) called *alpha particles* (α):



All of these spontaneous methods of restoring a nucleus to a stable configuration—known as *decay* processes—also involve the release of energy. The energy can take many forms. Ejected particles have a certain amount of kinetic energy but heat and even light may be released by highly radioactive materials. The ejection of charged particles like beta and alpha particles sometimes leaves the resulting new nucleus (the *daughter* nucleus) with excess energy. This state is in some ways analogous to an excited electron state for an atom. The transition of the excited nucleus to a lower energy state is generally accompanied by the emission of electromagnetic radiation of very short wavelength, *gamma rays* (γ). Such decays are regulated by whatever is going on in the nucleus and indeed gamma ray spectra for radioisotopes can be used for identification purposes.

The energies of the radioactive emissions as well as their makeup determine their interaction with matter. Because gamma rays are electromagnetic radiation and typically have considerable energies, they are generally able to penetrate solid matter some distance. Compared to alpha particles, beta particles have a miniscule mass and a small charge so they have the next greatest penetrating power. Larger and more highly charged alpha particles have problems getting through a piece of ordinary paper. Simple experiments can be done to sort isotopes by their decay mode by comparing the type of shielding (paper, plastic, metal, etc.) needed to cut down the intensity of the radiation by a fixed percentage.

The decay of radioactive materials follows first order kinetics, i.e., the rate of decay depends on the amount of sample present. The integrated form of the first order kinetics equation shows a logarithmic decrease in the amount of parent nucleus over time:

$$\ln N = -kt + \ln N_0$$
$$y = mx + b$$

where N is the amount of sample at time t , N_0 is the amount of sample present at $t=0$, and k is the rate constant for the decay. A useful benchmark value for comparing decay rates is the quantity known as the *half-life* for the process ($t_{1/2}$). This is the time it takes for half of the parent nuclei to change into daughter nuclei. The relationship between the half-life and the rate constant can be found by inserting a $N_0:N$ ratio of 2:1 in the integrated rate law:

$$\ln 1 = -kt + \ln 2$$

$$0.693 = kt_{1/2}$$

The half-lives of radioactive isotopes vary widely, with the most stable having values in the billions of years and the least stable in fractions of seconds. Since each decay event is typically responsible for the release of one particle or photon (in the case of gamma emission), it is possible to follow the kinetics of some decays by counting the number of particles ejected during a series of time intervals using an appropriate detector. Such a method, however, is not practical for isotopes with very long or very short half-lives. In these cases it must be possible to quantify the sample very precisely.

Since the rate of decay is directly proportional to the number of nuclei present, the following must be true:

$$\text{rate} = kN$$

where N is the number of nuclei present in the sample. The “rate” may be expressed in a variety of ways. A common unit for the activity of a sample is the Curie (Ci), 3.7×10^{10} disintegrations/s, or the microCurie (μCi), 37000 dps. After some statistical massaging the counts from a Geiger counter may be interpreted as disintegrations/time. Thus by repeatedly counting an accurately quantified sample of an isotope with a long half-life it is possible to calculate the rate constant, k , and so determine the half-life.

There are two common instruments for detecting/counting decay events. The most familiar is the *Geiger counter*. The detecting apparatus in a Geiger counter is the Geiger-Müller tube. The tube consists of a metal cylinder closed at both ends (cathode) with a thin metal wire (anode) suspended along the center axis. At one end or on the side of the cylinder there is a thin and fragile "window" which permits the passage of small charged particles and high energy photons (like gamma rays):

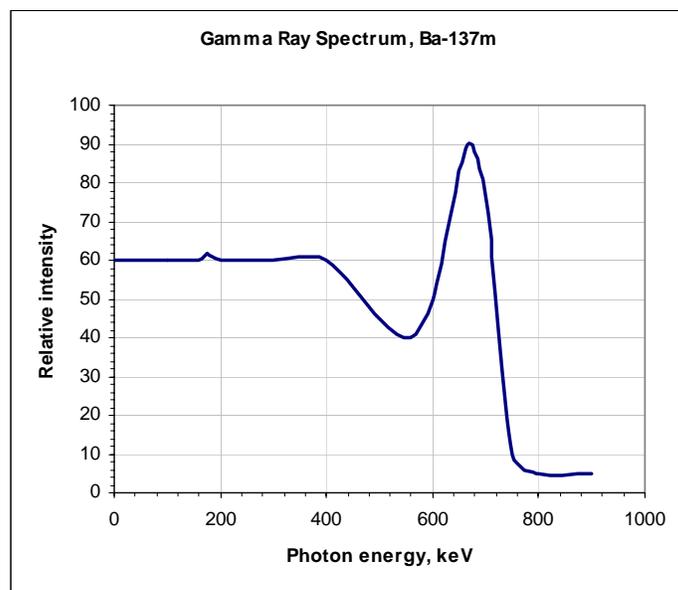
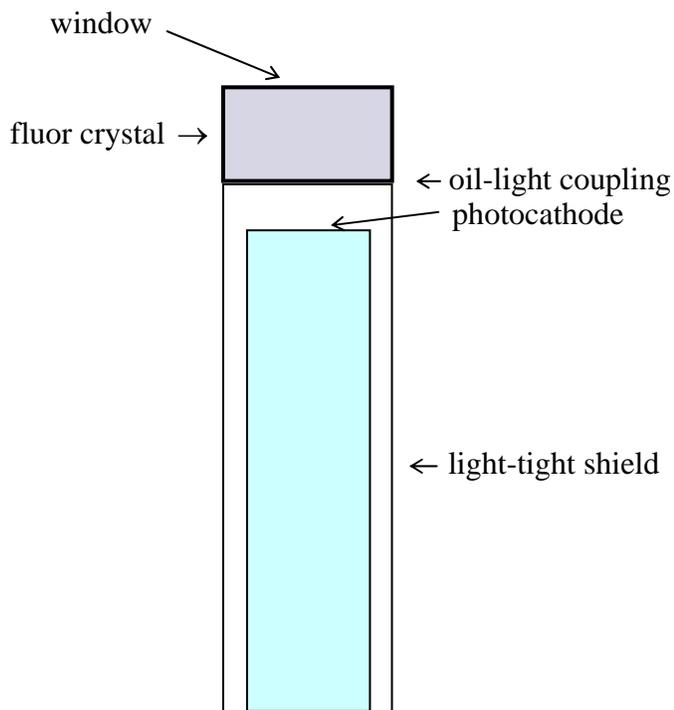


The tube is filled with argon gas at low pressure and has a high voltage applied across the anode and cathode. When charged particles or high energy photons enter the window they can ionize the gas atoms, creating more charges which move between the anode and cathode and temporarily allow an electric current to flow. This current is registered by the supporting electronics of the counter and may produce an audible or visible signal, or be used to drive a counting device. Each tube has a characteristic "threshold" operating voltage which must be appropriately set before use.

Geiger-Müller tubes generally contain a second gas which is present to "quench" the ion cascade that results from the entry of a charged particle or photon. Even so, there is a measurable amount of "dead time" during counting. Statistical methods for treating data to compensate for this limitation are required for experiments of significant duration and for accurate work where quantification of a sample is important.

Unless the “window” is very thin, most alpha particles cannot get through. Beta particles are easily detected but gamma rays may pass right through the tube without effect if their energy is high. A typical tube may detect only 2% of gamma rays emitted by a sample. This is yet another reason why statistical adjustments must be made to data for all but the simplest experiments.

A second common type of detector is sensitive specifically to gamma radiation. The short wavelengths of gamma rays interact with some crystalline materials known as *fluors* to produce small flashes of visible light, or *scintillations*, with intensities proportional to the energy of the gamma ray photon. A common fluor material is sodium iodide with a small amount of thallium present. Scintillations can be detected by photomultipliers which take the signal from a single flash and multiply it by a factor as high as 1 million. In addition to counting gamma events, scintillation detectors can be used to determine the specific energies of gamma photons and thus a kind of "gamma ray spectrum" for a radioactive isotope.



Although nuclear decays follow first order kinetics the decay of an individual nucleus is essentially a random event within some broad probability parameters. This fact, coupled with the nature of detecting devices like the Geiger counter, makes a better understanding of some of the statistics behind decay processes and counting helpful. Fortunately the principles are reasonably straightforward and in some cases the actual statistical treatments can be ignored if certain experimental parameters are tightly controlled.

The simplest "adjustment" to the count obtained from a Geiger counter is a subtraction of the "background count". Since experiments take place in the natural environment in which there are infrequent but detectable decay events, some of these events will be recorded along with any sample count. It is especially important to eliminate these stray decay events when quantifying a sample or determining a half-life. For this reason it is customary to take background counts before or after a sample count and subtract these counts (for a fixed period of time) from the sample counts before any further operations or adjustments are made.

It has already been stated that decay events are somewhat random. A better appreciation of what this means may come from realizing that the rate constant, k , is actually an expression of *the probability for a certain nucleus to decay per unit time*. For example, Cs-137 has a half-life of 30 years. Expressed in minutes this value would be 1.6×10^7 minutes. Calculating the corresponding rate constant ($0.693 = kt_{1/2}$) we have $4.4 \times 10^{-8}/\text{min}$. This number means that if you wait around for a minute and watch any Cs-137 nucleus, the probability that it will decay is 4.4×10^{-8} or 1 in 23 million! While this value may seem to place an actual decay event in the realm of the “not very likely”, even a 1 μCi sample of Cs-137 makes quite a racket when placed near a Geiger counter. Numbers of these magnitudes can be very deceptive.

A 1 μCi sample of anything would have a disintegration rate of 37,000 dps. The half-life of the Cs-137 in seconds would be 9.5×10^8 s and a corresponding k would be $7.3 \times 10^{-10}/\text{s}$.

$$\text{rate} = kN = 37,000 \text{ dps} = (7.3 \times 10^{-10}/\text{s}) N \quad \text{or} \quad N = 5.1 \times 10^{13} \text{ nuclei}$$

This is a very large number of nuclei, even compared to 23 million, so there are plenty of decay events in the sample each minute even with a half-life of 30 years.

With this probability consideration comes a certain statistical uncertainty in every count. Without going into a deep discussion of various statistical distribution models, this uncertainty is generally expressed as:

$$n \pm \sqrt{n}$$

where n is the number of counts (not counts/time). For example, if a sample gives 102 counts in a 10 minute period, this would be expressed as $102 \pm \sqrt{102}$ or 102 ± 10 counts.

Expressed as counts/min (cpm) this would then be 10 ± 1.0 cpm. This indicates about a 10% uncertainty ($1.0/10$)—or 90% accuracy—in the count. Clearly, more counts tend to lower the *relative* statistical uncertainty just as flipping a coin 1000 times as opposed to 10 times tends to approach the 50-50 chance of heads or tails more convincingly.

So how many counts is “enough”? This depends on at least two factors: the count rate itself and the desired accuracy. To make a decision it is helpful to keep in mind the expression for the fractional statistical uncertainty in the count: $1/\sqrt{n}$. To have a count with a 90% accuracy or 10% uncertainty means $0.10 = 1/\sqrt{n}$ or $n = 100$ (similar to the original example). If you want to increase the accuracy to 95% (now the statistical uncertainty would be only 5%) then $n = 400$! In practical terms, if a sample gave a count rate of about 40 cpm, you would need to count for at least 10 minutes ($10 \times 40 = 400$ counts) to reduce the statistical uncertainty to 5%.

As a routine matter this kind of thing can get tedious but some of the burden is removed by the software that runs the Geiger counters used in the lab. Because of the limitations of the electronics involved, the instruments count decay events until a pre-set total has been measured. The time required for this count is then used to calculate and display a count rate (cpm). The statistical uncertainty is displayed along with this value so it is easy to see if the accuracy is sufficient and, if not, to increase the number of counts accordingly. For our experiments we will aim for a maximum of 10% uncertainty. The background count time period has been set to deliver this kind of accuracy for our locale but sample counts will vary depending on the activity of the isotope, size and geometry of the sample.

Sample geometry introduces yet another variable. In the simplest cases where quantification is not an issue, this can generally be ignored. Where it cannot be ignored and the sample is essentially a point source (or a drop of solution), reproducing the geometry as accurately as possible for each sample is a “low tech” mechanical approach to dealing with the problem. A simple numerical approach assumes that half of the count is missed because half of the radiation is headed toward the countertop instead of the Geiger tube! Thus doubling the measured count “corrects” for this geometric problem.

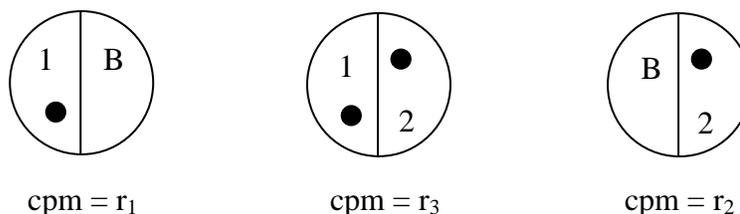
In reality more than half the decay events may be “missed”. Geiger tubes have varying efficiencies for detecting radiations of different energies. A Geiger tube which is 80% efficient for a 1 MeV (mega electron volt) gamma ray will only indicate 4 out of every 5 events with that energy (4/5 = 0.8). The fifth gamma ray may pass right through the tube without initiating a signal event. When samples of the same substance and comparable activities and geometries are compared (but not quantified), the efficiency correction can also be generally ignored. When this is not possible, a simple combined correction for background, efficiency and geometry might look like this:

$$\text{actual dpm} = (\text{cpm} - \text{bkgrd}) \times \frac{1 \text{ dpm}}{0.\text{eff}(\text{cpm})} \times 2$$

where **dpm** is the disintegrations/min, **cpm** is the measured count/min, **bkgrd** is the background count for the same time period, and **0.eff** is the decimal efficiency for the observed decay and the Geiger tube used. The factor of “2” is the simple geometry correction.

Finally, Geiger tubes have an inherent “dead time” while positive ions travel to the cathode and another signal cannot be recorded during this time. A typical correction for this *resolving time* (sounds better than “dead time” in a brochure) is 0.5% per 1000 cpm based on an assumed resolving time of about 300 μs. For samples with activities lower than 1000 cpm the correction is negligible.

Special isotope samples are manufactured for the determination of resolving time. They typically consist of a disk which is divided in half. Each half contains an “identical” sample of Tl-204. A “blank” half disk without the isotope is also provided. The geometry of the various pieces for a series of three counts looks like this:



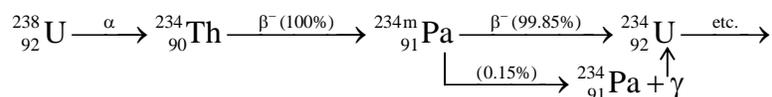
$$\text{resolving time (in minutes)} = T = \frac{r_1 + r_2 - r_3}{2r_1r_2}$$

$$\text{corrected activity} = \frac{\text{observed activity}}{1 - (\text{observed activity})T}$$

Extended decay sequences are very common for heavy elements like uranium, and as samples of uranium compounds age they will become increasingly “contaminated” by small amounts of various daughter isotopes. In some cases these products can be easily extracted and studied in the lab. In the case of U-238, there are 13 steps in a decay sequence which ultimately ends in lead-206.

[In addition to this “natural” decay sequence (i.e., it occurs in the natural world without human initiation or intervention) there are three other sequences generally cited as “natural”. They include the decay of uranium-235 to lead-207 (sometimes called the “actinium series”), thorium-232 to lead-208, and neptunium-237 to bismuth-209. The last series no longer occurs naturally on this planet owing to the shorter half-life of neptunium, its presumed initial abundance, and the age of the earth. Synthesized samples of Np-237 would still follow this decay sequence, however.]

One of the early daughter isotopes in the U-238 series is Pa-234m. The “m” stands for *metastable* and indicates a nucleus with additional energy. The formation of such nuclei follows statistical rules and the decay of the metastable forms is generally accomplished by the release of a gamma ray of wavelength corresponding to the energy difference between the metastable and regular form. This is known as an *isomeric transition* (IT). The variables behind the kinetics of gamma ray emission are related to quantum spin states and the angular momenta of the parent and daughter nuclei. Some gamma emissions are so fast (less than nanoseconds) they are virtually simultaneous with charged particle emission. Those nuclei with gamma emission half-lives larger than 1 nanosecond (10^{-9} second) are said to be metastable.



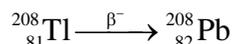
In this case it is not the isomeric transition itself that we follow (a half-life of about 6.7 hrs) but rather the majority faster and direct beta decays of the metastable nuclei to uranium-234.

The separation of Pa-234m from the uranium and its other daughter isotopes so that it can be counted turns out to be relatively simple. An acidified solution of about 1 g of uranyl nitrate in 15 mL of 7 M HCl is vigorously shaken with an equal volume of 4-methyl-2-pentanone in which the Pa(V) cations are more soluble than in the acidic aqueous phase. The ketone layer can then be counted as the Pa-234m decays (a lead shield covers the lower aqueous layer). The entire extraction process takes place in sealed polyethylene bottle so there is never any direct contact with the isotopes. The Pa “generator” can be reused within an hour and will last for many years.

In the thorium decay sequence there are several relatively short-lived isotopes near the end which can be easily isolated chemically and used for half-life determination. One is bismuth-212 which will spontaneously plate onto a more active metal sample such as nickel when it is placed in an “old” thorium solution. The half-life for this isotope is on the order of 60 minutes and small samples can be difficult to count because of the low activity.

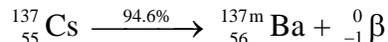
[An “old” solution is one in which the daughter products have reached what is known as *secular equilibrium* with the parent isotope; this happens with a long-lived parent and short-lived daughters and results in a mixture in which the daughters eventually decay at the same rate they are produced, i.e., the parent and daughter will decay at the same rate until the parent is exhausted. One way to obtain an “old” thorium solution is to extract thorium nitrate from old lantern mantles. At one time these little silk pouches that are still used today for camping lanterns were dipped in thorium nitrate to increase the brightness of the lantern light (about 300 mg Th per mantle). When the mantles are initially ignited, the silk burns away leaving a fragile ceramic oxide structure which, when heated by the burning gas, give off an intense light. The phenomenon has nothing to do with radioactivity but instead a shared property of a number of metal oxides and is related to the bright “limelight” lamps used in early English theatre in which heated calcium oxide (“lime”) produced a very bright light. Most mantles manufactured today in this country are treated with yttrium nitrate instead but some mantles from overseas are still treated with thorium compounds. However, newer mantles have very small concentration of daughters present.]

At the end of the same thorium decay sequence, thallium-208 has a much shorter half-life:

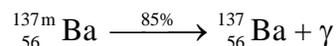


It can be removed from a solution by *adsorption* onto a bed of precipitate. This is one method sometimes used to separate tiny amounts of isotopes from a solution. In our experiment an ammonium phosphomolybdate precipitate is used to trap the thallium isotope for counting as the thorium solution is poured through it (the filtrate is removed prior to the washing step for potential reuse in about 30 minutes).

Still another isotope that may be studied for its decay kinetics in this experiment is a metastable form of Ba-137. It is produced by the beta decay of the long-lived isotope, Cs-137:



[A small number of Cs-137 nuclei (5.4%) decay directly to the stable form of Ba-137] The half-life for this decay is 30.17 years. The beta decay is detectable but slow for use in a laboratory half-life determination. However, the metastable Ba-137 undergoes an isomeric transition by emitting a gamma ray. The Ba-137m nucleus has a half-life within the range of practical measurements in the laboratory:



The determination of the half-life of an isotope in the student laboratory is a moderately interesting activity if only because of the sheer novelty of working with radioactive material. The application of measurements like this to answer questions about the age of a sample or artifact is significantly more interesting. Items which lend themselves to this kind of investigation are usually mineral specimens of some kind or archeological artifacts of organic origin. In these samples there must be trace amounts of long-lived radioisotope parents and non-radioactive daughters. Knowing the half-life for the decay, it is possible to compare the *current* amount of parent (N) + daughter in the sample and take this as the *starting* amount of parent (N_0 in the integrated rate law seen earlier). A calculation of the time elapsed is “simple” but extraction of a representative sample and accurate quantification of very small amounts is not.

Although such determinations are generally outside the range of possibilities for introductory teaching labs, it is still possible to “date” appropriately chosen samples with simple equipment. Tiny license-exempt samples of relatively short-lived isotopes are commercially available in sealed plastic discs for teaching purposes. The available half-lives of typical samples range from 30 years to about 6 months. With an aging inventory of samples purchased over a number of years it is possible to compare the activity of a “new” sample with one that is considerably older. With careful control of sample geometry and a correction for the loss of activity in the “new” sample since manufacture, a reasonably good value for the age of the older sample is possible.

Finally, much radiochemical work requires careful quantification, something already made clear in the previous discussion. The ubiquity of radioisotopes in the natural environment (albeit hopefully in very small amounts!) is something that many students fail to grasp. We carry carbon-14 in our bodies all the time, of course, but radon gas also percolates up through the soil from decaying isotopes in the earth. Potassium—which is naturally concentrated in some foods like apricots and bananas—has a radioactive isotope (γ emitter) with a very small abundance (0.0117%). Even so, bottles of potassium chloride from the stockroom shelf give a count measurably above the normal background.

This latter case is particularly interesting because of the food angle. Of course, one would have to eat enormous amounts of food containing potassium to ingest a tiny amount of the radioactive potassium (K-40), but it *is* there. Potassium chloride is also available in a commercial salt substitute manufactured for people who must limit their intake of sodium. Morton’s Lite Salt™ contains a significant concentration of KCl mixed with NaCl. The activity is quite low and it is an interesting exercise in counting statistics and experimental precision to attempt to determine the K-40 content of the mixture. The use of a fairly concentrated granular sample eliminates most of the sample geometry concerns that would arise from attempting to count dried apricots or bananas. The process can also be simplified quite a bit by using a “known”, i.e., a sample of reagent grade KCl, for a standard. Equal masses of the pure KCl and the Lite Salt™ in essentially identical geometries can be counted and the activities (proportional to the K-40 content) compared.

The Experiment

There are five parts to this experiment:

- investigation of shielding materials for alpha, beta and gamma radiation
- determination of resolving time for the Geiger counter
- determination of the age of commercial isotope samples
- quantification of K-40 in Lite Salt™
- determination of the half-life of a selected isotope

The following non-locker materials will be provided:

- paper and aluminum pieces
- assorted isotope disc sources of various ages
- Tl-204 resolving time isotope set
- Pa-234m isotope generators w/lead shield
- Cs-137/Ba-137m isotope generator
- Eluting solution (0.9% NaCl in 0.04 M HCl)
- steel planchets
- Beta-shield gloves
- “old” Th-232 solution
- acidified 0.78 M NH_4NO_3
- 0.065 M $\text{NH}_4\text{H}_2\text{PO}_4$
- 0.086 M $(\text{NH}_4)_6\text{Mo}_7\text{O}_{24}$
- 3 M HNO_3
- microscale suction filtration apparatus
- Geiger counters w/CBL interfaces

The Chemicals

Uranyl nitrate, $\text{UO}_2(\text{NO}_3)_2 \cdot 6 \text{H}_2\text{O}$, is a yellow crystalline solid. The crystals show a marked tendency to display triboluminescent behavior when shaken, rubbed or crushed, with occasional detonations. The salt has been used in a glaze for decorating porcelain.

4-methyl-2-pentanone (also known as isopropylacetone and methyl isobutyl ketone) is a colorless liquid with a faint odor. It is used as a solvent for gums, resins, nitrocellulose, etc. It may be irritating to eyes, mucous membranes and narcotic in high concentrations.

Thorium nitrate, $\text{Th}(\text{NO}_3)_4 \cdot 4 \text{H}_2\text{O}$, can be obtained from monazite sands. It consists of white, deliquescent crystals. This compound with a 1% cerium nitrate additive constitutes the usual impregnating liquid for incandescent mantles. It has been used in the past to treat parasitic skin diseases.

Ammonium nitrate exists as odorless, colorless crystals or white granules. It is used for making dinitrogen monoxide (“laughing gas”), in freezing mixtures, matches, pyrotechnics and fertilizers. Disastrous explosions attributed to ammonium nitrate have occurred from time to time, the most notable in recent history being the accidental one at Texas City and the intentional attack on the Federal Building at Oklahoma City in 1995.

Ammonium dihydrogen phosphate (or monobasic ammonium phosphate) consists of odorless crystals or a white crystalline powder. It has been used as a baking powder with sodium hydrogen carbonate, in fermentation mixtures and for the fireproofing of paper and wood.

Ammonium molybdate is either colorless or slightly greenish or yellowish. It is used in photography, for decorating ceramics and commonly for the determination of phosphate in the analytical lab.

Nitric acid has been called "aqua fortis" (strong water). It is generally produced by the oxidation of ammonia followed by reaction of the gaseous products with water. When pure it is a colorless liquid that fumes in air with a characteristic choking odor. "Concentrated" nitric acid is a water solution containing 70% HNO_3 . Even dilute solutions will stain woolen fabrics and animal tissue yellow. It is a very strong oxidizing agent, reacting violently with most organic matter.

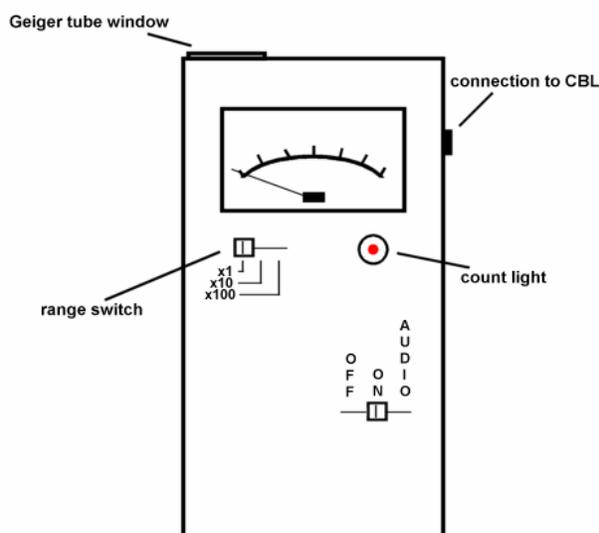
Nitric acid is used in the manufacture of fertilizers, dye intermediates and explosives.

The *eluting solution* in this experiment contains only dilute NaCl and HCl. It presents no special hazard. The Ba-137m *isotope generator* itself contains radioactive Cs-137 bound to an ion exchange resin. The amount of isotope is very small (less than 10 μCi [1 Ci--or Curie--equals 3.70×10^{10} disintegrations per second]) and license-exempt. The generator is sealed except for the column inlet and outlet. There *is* detectable low-level beta radiation emitted from the generator but these particles are not particularly energetic. Nonetheless, beta-shield gloves (lead lined) should be worn when handling the generator. **To minimize the potential for contamination, the instructor will dispense the Ba-137m isotope.** After about 30 minutes following elution, the activity of the solution is less than 1/1000 of its original value and the small amount of material represents no hazard.

Technique Discussion

[this experiment will be done with a partner; because of the limited number of samples it may not be possible for all students to do the same part of the experiment simultaneously]

The Geiger counters which attach to the CBLs are relatively simple devices. The tube itself is completely enclosed within the counter case. When ionizing radiation enters the tube there is an indication by a flashing light, a movement in the meter and an optional audio signal [no one in the lab wants to listen to ten Geiger counters—do not use the "Audio" setting]. A signal is also sent to the CBL, of course, and the calculator program can interpret this data in a variety of ways. The meter itself is only useful for rough approximations or surveys but it can be damaged by high levels of activity if the range switch is not properly set. Always check to see if a sample causes the meter reading to move consistently off scale. If it does, increase the range setting to x10 or x100. This will not affect the data sent to the CBL.



The CBL Geiger counters will be provided clamped on a ring stand in a generally useful position (upside down, window pointing toward the lab bench). If you need to move the counter (for the half-life determination of Pa-234m) be careful when retightening the clamp so that you do not break the case. Observe the original position of the clamp before removing it. Remember, counting geometry is very important for many parts of the experiment in order to achieve consistent results. You can trace the outline of a disc source on a piece of paper and set the ring stand on the paper, positioning the counter window directly over the circle. This may help to control some geometric variables.

The different parts of the experiment do not need to be done in any particular order, i.e., there are no sequences in which later work depends upon earlier work. You should proceed through the different parts as samples become available OR as assigned by the instructor. *Remember that all counts should have relative uncertainties of 10% or less.* If they do not you will need to increase the number of counts until they do.

The program that runs the Geiger counter/CBL from your calculator is fairly simple to use. **Be sure to bring your TI-83/P calculator to lab and have the GCOUNTER.83G program in memory.** There are only 3 main options for the software: *background count*, *ratemeter*, and *half-life*. Both the background count and ratemeter default to 100 counts. In this area such a *background count* should take about 7 minutes and come very close to the 10% uncertainty. After the background count is concluded you are given the option to adjust subsequent counts for the correct factor or not adjust them. In general, you will want to make the adjustment (but record the background count anyway).

The *ratemeter* function is designed to function like any other counter/ratemeter except that you cannot count for a set time. Instead, you select a number of counts (default=100) and the CBL counts until it reaches that value, then divides by the time it took and displays count/min along with the uncertainty. You have the option to take a count only once or for up to 5 times in succession. If the uncertainty for a count is too high you can change the default number of counts from a selection at the starting menu.

The *half-life* function is designed for relatively short-lived isotopes and you will need to specify the approximate period of time you want to count. Rates in cpm are scrolled up the calculator screen in real time and stored in the calculator lists.

The investigation of shielding materials may be done with any combination of alpha, beta and gamma disc sources. Background counts are not necessary for this part of the experiment. After determining distances at which the three samples give approximately equal apparent activities (~ 100 cpm using the survey meter), try the paper and aluminum pieces to determine which emanations are stopped (or significantly diminished) by which materials. Always count the **back** sides (non-printed) of the discs. According to the supplier, detection of activity may occur through the back of the Geiger counter case with gamma rays or high energy beta particles. If aluminum metal (about 1/8") is placed between the sample and the case and the activity drops dramatically, the radiation is most likely beta. For weaker beta samples and also alpha emitters, the end window should be used. A sheet of paper inserted between the sample and the window should eliminate most alpha detection.

The resolving time determination is done according to the description given in the **Background** section. After taking a background count have the calculator correct subsequent counts to compensate. Place the samples about 1/2" from the counter window and obtain *three* 150 count samples for each of the three configurations. The manufacturers marks should face **up**.

You will be assigned an isotope disc sample to “date”. After taking a background count (choose to adjust subsequent counts) place the “new” sample, back side “up”, about ½ inch from the counter window. Adjust the counter calculator program for a 500 count sample (as opposed to the default 100 count sample). Count the sample 5 times without moving it. Record the date stamped on the “new” sample. Replace the “new” sample with the old one (back side “up” **unless** otherwise indicated), trying to carefully reproduce the counting geometry. Count the sample 1 time without moving it.

For the determination of the KCl content of Lite Salt™ adjust the calculator program for a 200 count sample. After taking a background count (choose to adjust subsequent counts) measure 8.00 g of KCl into the small metal sample holder, level it, and count with the counter touching the rim of the sample holder. Return the KCl to the stock bottle. Clean out the sample holder thoroughly and measure out 8.00 g of Lite Salt™. Level the sample and count as before. The data for these counts should be submitted to the instructor before you leave the lab so that a class set may be distributed.

There are three different isotopes available for the half-life determination. You will be assigned one. The general procedure is to first take a background count (opt to adjust later counts), adjust the sample count as per the instructions for your isotope, adjust the counter itself to bring it within the distance specified and then count for the recommended time. Follow any other particular instructions for your specific isotope, especially those pertaining to safe handling and disposal.

Ba-137m: Adjust the calculator program for a 250 count sample. Position the Geiger counter window ½ inch above the lab bench. The instructor will force a few drops of eluting solution through the ion exchange column on which the Cs-137 parent isotope is fixed. The eluted Ba-137m daughter will be collected in a small stainless steel dish called a *planchet*. Counting should commence immediately for 10 minutes. After the count is complete the sample should be removed to the fume hood for eventual disposal.

Pa-234m: Adjust the calculator program for a 100 count sample. Remove the Geiger counter from the clamp and place it on a few masonite squares so that the counting window is level with the ketone layer (upper) in the sealed Pa-234m generator. The lead shield should be wrapped around the lower part of the bottle where the acidic aqueous phase is. Place the counter in contact with the bottle and take a background count. Choose the option to adjust subsequent counts. Seal the bottle in a zip-lock bag and shake it vigorously for 30 seconds. Remove the bottle from the bag, replace it in the lead shield, allowing a total of 30 seconds to elapse. Begin counting for 7 minutes.

Tl-208: Adjust the calculator program for a 100 count sample and position the counter ½ inch above the lab bench. **You must wear gloves for the separation operation and you must work only on the plastic sheet that is taped onto the lab bench. To avoid contaminating a calculator or CBL, one student should work only with those items while the other student handles the radioactive materials [with gloves].** In a 100 mL beaker mix 8 mL of acidified NH_4NO_3 solution with 1 mL of $\text{NH}_4\text{H}_2\text{PO}_4$ solution. Add 3 mL of $(\text{NH}_4)_6\text{Mo}_7\text{O}_{24}$ solution and heat the mixture in a water bath at 80°C for 20 minutes. Use a small amount of water to seat a circle of paper in the bottom of the filtering crucible and transfer the warm solution to the filtering crucible using medium suction. A bed of yellow precipitate should deposit at the bottom of the crucible. Wash with several mL of distilled water. Remove the vacuum filtering flask and replace it with a clean dry flask. Add the sample of “old” thorium nitrate to the filter crucible slowly, dropwise, under low suction. **Remove the vacuum filtering flask containing the thorium solution and set it aside.** Quickly replace the first filtering flask and wash the precipitate *twice* with 10 mL portions of 3 M HNO_3 . Take less than a minute to “dry” the precipitate by drawing air through it and then immediately transfer the paper and precipitate to a small stainless steel dish called a *planchet* and begin counting for 10 minutes. During the counting process carefully return the initial filtrate to the “old” thorium nitrate bottle. Leave all of the equipment used for the separation on the plastic sheet. When the sample count is complete, remove the sample to the fume hood for eventual disposal. Carefully peel off the gloves (so the outside ends up on the inside) and add them to the used glassware on the sheet. Wash your hands!

The Report

Your initial calculations should include:

1. The resolving time for the Geiger counter you used (μs)
2. Time elapsed (i.e., age) since the manufacture of the “old” isotope disc
[assume stamped date on “new” sample is the 1st of that month and round fractional months]
3. Grams of K-40 in 8.00 g KCl
4. Apparent grams of K-40 in 8.00 g Lite SaltTM based on activity
5. Grams of KCl in 8.00 g Lite SaltTM
6. Average g of KCl/8.00 g Lite SaltTM based on class data and % KCl by mass in Lite SaltTM
[manufacturer states 46.3% by mass---give relative error]
7. A graph of $\ln(\text{counts})$ [corrected for background] vs. time (minutes) for assigned isotope
8. The rate constant and half-life for the isotope [relative error]

Include a brief discussion of the shielding experiment in your conclusion along with comments regarding any large deviations in your results.