

The apparent change of activity with temperature in a ^{226}Ra decay chain

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Received: 29 June 2011 / Published online: 31 August 2011
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Abstract Radioactive decay rates are to a large extent believed to be independent of the chemical environment. This is the physics basis implicitly assumed in applications such as radioisotope dating. While this statement is a good approximation for most radioactive decays, there are cases where a slight variation of 0.5% or more can be observed, as in the electron capture type of decay. There are renewed interests in possible decay-rate changes with external parameters such as temperature, with controversy as to the phenomenon's authenticity. In this paper, we study the variation of radioactivity *counts* that significantly change (up to 50% or more) with temperature. We carefully studied the characteristics of the change and found that the presence of a gaseous decay daughter can pose a serious challenge to a *bona fide* account of the intrinsic nuclear decay rate. After a careful solution to rate equations of the relevant isotopes under our experimental conditions, we found that most of the radioactivity change could be accounted for by the diffusion and loss of gaseous daughters under the heat, without a supposed change in the intrinsic nuclear decay rate. We hence demonstrate that an accurate determination of the decay constant has to consider the possible diffusion of volatile components in the

decay chain. This is especially important in cases involving significant temperature change.

Keywords Radioactive decay constant · Decay rate · Radioactivity · Chemical environment · Radium

Introduction

The rate of radioactive decay is often said to be independent of the chemical environment. However, some recent results suggest the possibility that decay rates might have a weak dependence (0.5% or less) on temperature [1–4]. In those experiments, the temperature T was typically lowered, resulting in a claimed acceleration in the decay rate. Such claimed changes are so far highly controversial, with many counter-claims giving null results within experimental errors [5–7].

However, it is well known in the field of radiochemistry that the apparent radioactivity count is affected by the possible migration of radioisotopes in the sample. Such a migration can be caused by chemical leaching, sorption and diffusion as reported before [8–11]. We will see later in this paper that the change in the apparent radioactivity can also be caused by diffusion of isotopes, effected by means such as heating.

Partly motivated by controversies outlined as above, we decided to do a test of our own. Instead of the well investigated isotopes like ^7Be , ^{198}Au , ^{22}Na , we decided to probe in the well-known decay chain of radium (^{226}Ra). Two significant differences to prior experiments are in the temperature and in situ heating of the isotope with an electrical power. Here we want to increase the temperature rather than to drop it. This gives potentially more room in the temperature control compared to the cryogenic

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approach where there is also a cryogenic limit at absolute zero. Our electrical control of input power also gives potentially more temperature control.

Experimental

We utilized some fairly porous and powdery materials for the experiment. The logic is as follows. Because we wanted to test how much the environment can have on the radioactivity, one obvious solution is to make ^{226}Ra as far away as possible from its ordinary solid-state form, where Ra atoms are closely packed one to another. One natural choice is to make the Ra in a very finely divided nanoporous-form, supported by a Pd metal matrix that is itself a fine-divided powder. In doing so we hope that radium atoms are in sufficient contact with gaseous elements when the powder is flushed with gases. The choice of Pd as the matrix material is due to its high affinity with hydrogen which in a hope to dramatically change the environment to the Ra. We utilize Pd also due to our familiarity with nanoscale Pd from our prior works in fuel cell catalysts [12–14], and energetic systems [15].

The ^{226}Ra is mixed on a nano-palladium black from a ^{226}Ra nitrate solution. The ^{226}Ra was ordered from Eckert & Ziegler Analytics and had a total activity of 1 μCi . The nano-palladium black was prepared from acidified palladium chloride in distilled water. Twenty grams of palladium chloride dissolved in 200 mL of distilled water gives a pH ~ 3 . The Pd black was reduced with 50 g of Zn metal shavings over a week, followed by decantation, settling with repeated rinsing, up to 10 times to remove zinc ions. It is then vacuum-dried over calcium chloride, resulted in an extremely fine reduced Pd black powder, termed nano-Pd thereafter.

The nano-Pd powder has a Stokes settling time of 2 weeks over a vertical distance of 2 cm. The settling velocity thus calculated enables us to estimate the average radius of the Pd particles to be 0.025 μm or 25 nm. The diameter of the nano-Pd particles are therefore on the order of 50 nm which is independently verified by scanning electron microscopy (SEM).

Then the radium nitrate solution of 2 μM (micro molar) was added to the nano-Pd powder, well mixed and dried. This radioactive powder was then heated at 150 $^{\circ}\text{C}$ in a hydrogen furnace for 1.5 h to reduce the divalent Ra ions back to the metallic form. The sample was then cooled and flushed with nitrogen prior to removal.

The ^{226}Ra coated nano-Pd powder is mixed with a ZrO_2 powder (5–10 μm) at a ratio of 1:1.4 to finely adjust its electrical resistance. The resistance will later form the in situ heating of the powder when driven by an electrical current. The second function of the zirconia powder is to

further increase the interfacial area that the ^{226}Ra /nano-Pd powder can engage so as to further increase the environmental change to the ^{226}Ra . The ZrO_2 further serves as a test ingredient to see whether its presence can stop the diffusion of gaseous radioactive decay products, which in this case, are basically the radon gas.

The mixed radioactive powder is then loaded in the test cell shown in Fig. 1, which basically consists of a glass (Vycor) tube 1.0 cm in its inner diameter and measures 3.5 cm in length. The core temperature of the cell was either measured by three thermocouples attached to the outside of the glass tube or by an infrared pyrometer. The Vycor tube contains typically 2 g of radioactive ^{226}Ra /nano-Pd/ ZrO_2 powder in its central cavity, whose activity is counted by a gamma spectrometer probe placed ~ 2 cm from its outer glass wall. The ^{226}Ra /nano-Pd/ ZrO_2 powder in the cell chamber is driven by a DC electrical power source, and typically heated to ~ 120 $^{\circ}\text{C}$. A typical heating to this temperature is achieved with 1.9 V and 2.8 A with a power of 5.1 W. These electrical parameters are only a rule of thumb, because resistance of the radioactive powder changes with loading of hydrogen and expansion of the Pd lattice. Two discs made of nickel foam (open cell) clamp the powders in place in the Vycor tube. They also form the electric contacts with both the powder and the stainless steel (SS) tubes so that an effective electric contact with the DC power source can be carried out. A typical experiment starts with an activation phase, when different test gas filling the powder within the interstitial spaces, while the DC electrical power was applied. The gas pressure can be controlled between 1 psi (pound per square-inch) and 20 psi as a static system. The gas system is not necessarily 100% gas tight and therefore there are pressure swings (typically a drop) when the test was carried out. The gas flow is regulated by valves (not shown in Fig. 1) and typically this happens only in the activation stage.

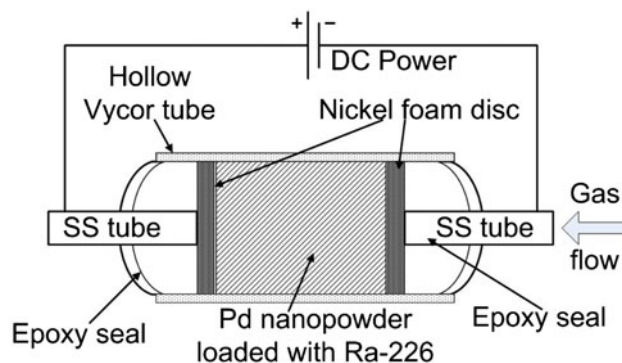


Fig. 1 The detail of the radioactive capsule. The major construct starts with a hollow Vycor tube, filled with radioactive nano-Pd powder which is then sealed in two sides by Devcon 5-min epoxy

Afterwards the valves are sealed off so there is not a forced gas flow when the radioactivity counting is carried out.

Different gases were utilized in the experiment, with the hope of achieving different inter-Ra interstitial environment so as to observe any potential activity change. The gases tested include hydrogen H_2 , deuterium D_2 , nitrogen N_2 and air. The former two are especially effective because they permeate the Pd matrices very well, and are well-known hydride formers for the alkaline earth group to which the Ra belongs. In fact the hydrogen isotopes give the largest activity drop observed and we will try to explain this later on.

The ^{226}Ra α -decays into ^{222}Rn . However the α -particles are relatively difficult to observe because of their large attenuation through the glass (Vycor) wall. The alphas are also too easily attenuated in the $^{226}\text{Ra}/\text{nano-Pd}/\text{ZrO}_2$ powder sample because we need a minimal thickness to maintain a suitable heating and temperature measurement. The alphas are typically detected in a surface-sensitive surface barrier detector (SBD) but there does not appear a convenient method to put the SBD in the heated tube without damaging it. If the α -particle is to be detected with a scintillation method, there comes another problem. The γ -rays from the daughter products (e.g., ^{214}Pb , ^{214}Bi) definitely pose an interference via their high scintillation efficiency. All these considerations explain why we finally decided to utilize gamma spectroscopy from the daughters of the ^{226}Ra decay chain to characterize its activity. The adoption of gamma spectroscopy makes the counting of activity a feasible task in the current experimental conditions.

We have utilized three different gamma instruments to observe the count rate. All three are independently calibrated first and cross-calibrated later on during the experiment. Gamma counts were in one way obtained using a Technical Associates (Canoga Park, CA) Ratemeter/Scaler Model # PRS-5 and Probe model # BGS 251 which utilizes a 1 inch thick BGO (bismuth germanate, $\text{Bi}_4\text{Ge}_3\text{O}_{12}$) crystal. Integration of counts was performed for 30, 60 and 90 min periods, over days. Gamma spectra were obtained in two ways. The first used a Radiation Solutions Inc. ^{125}RS . The ^{125}RS also counts the activity as well. The second and the most sensitive gamma spectroscopy is based on the ORTEC Model 276 photomultiplier with a preamplifier mated to a NaI scintillating crystal, powered by an EG&G 556 HV power rack. The main amplifier is the 590A, supported by an 871 timer/counter. The multi-channel analysis function is implemented by the ORTEC Maestro software package. The Maestro package also doubles as a count meter with some configurations in its job file. The ^{125}RS is self calibrated with natural gamma sources like potassium ^{40}K , uranium and thorium isotopes. The ORTEC spectrometer is triple calibrated with ^{137}Cs 662 keV and ^{60}Co (1.17 and 1.33 MeV) standards.

Of all the samples tested, roughly half showed significant drop ($>10\%$) with the heating ($\sim 125^\circ\text{C}$) within 10 h of experiments. The drops on the order of 5% are taken as inconclusive and will not be discussed here. We found out that the samples with significant drops typically happened after the radioactive powders were stored in a sealed condition for a long time (1 month or so), and had a large count rate to start with, say over 30,000 CPM with the Technical Associates Model # PRS-5 counter. We call this kind of powder a “hot-shot”. In comparison, those samples that were fresh prepared or had gone through an extended period (say a few days) of heating and gas flushing procedures immediately before the test typically would not show much change. This observation serves an important hint to the real cause of the activity drop and is well explained with our theoretical model. This is to be detailed later part of the paper.

Of course there exist concerns as to whether or not the observed change in the count is in fact due to the temperature change of the scintillation detectors. The light yields of all of the scintillators (BGO, NaI) that are utilized in the study are highly susceptible to temperature change. If the detectors’ temperature was affected by the test capsule’s temperature while the gamma counting was based on simple discrimination methods, the count rate will also change as outlined above. However, we will show later in the paper that this is not the case. For now we focus on the essential results so that the discussion of detector temperature and the test to probe it is deferred to the section entitled “Possible temperature change in the detector”.

There are small statistical fluctuations from counts in 1 min to those in the next one. However, the fluctuation is never over 2% on a counts-per-minute (CPM) basis with the Ortec spectrometer. Therefore, if there are a long term change of counts in the range of 10% or more, we cannot attribute it to a statistical fluctuation. A typical test run of the count drop with deuterium is given in Fig. 2. The total drop is over 50% over 7 h. To check if the drop in radioactivity is specific to the particular hydrogen isotope of deuterium, we also ran the test with light hydrogen. The results were positive as well as given by the upper part of Fig. 2. The experiment with deuterium in Fig. 2 was carried out in two stages with a total drop of 41%. Only the second part of drop ($\sim 21\%$) is given in Fig. 2.

Similar tests were carried out with vacuum, air and nitrogen. All “hot-shot” powders showed some drop of radioactivity, typically on the order of 15% or more. The drops with vacuum, air and nitrogen were typically less than cases with hydrogen isotopes, not significantly more than 20%. We tentatively attribute this percentage difference in the activity drop to the permeable nature of hydrogen, although an in-depth research on this is desired. More quantitative results on the two gases and vacuum will

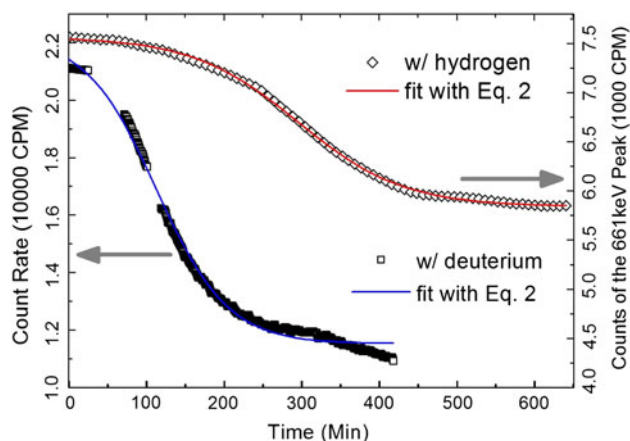


Fig. 2 The drop of radioactivity in the ^{226}Ra chain with deuterium/hydrogen permeating the sample electrically driven to increase its temperature. The discrete squares and diamonds are from the experiment. The blue and red lines are fits to the experimental data with the last formula in Eq. 2

be published elsewhere. It is not included here however due to the page limit. In all cases, the drop often happens in a few hours and then it slows down. In all tests the activity eventually recovered to its original value, if the heating current was turned off. However the recovery is much slower than the drop. It often takes a few days and up to 2 weeks to fully recover.

In many of the experiments discussed, the reading from the three spectrometers are cross-checked, sometimes also compare to the readings from additional Geiger counters. In all cases, the drops were observed by all detectors presented. The average change read from different instruments typically agrees with each other within 2%. This rules out any possible problem with the instrument.

With the phenomenon confirmed, a logical deduction is needed to explain it. Clearly merely mixing the nanoparticles together or applying an electric current via the valence electronic bands, it is still impossible to change the nuclear force. Also replacing Rd nuclei with hydrogen ones tend to reduce the average nuclear field around the decay Rd, and this should speed up the decay process according to quantum mechanics. This is in direct contradiction to the experimental results: the experiments always show a lowering in the activity with heating. Therefore, we can basically rule out this scenario, for now.

The other option is not to focus on the intrinsic decay rate from the ^{226}Ra , but rather on the activity of the radioactive decay chain. We notice that the gamma count is not directly associated with the rate of ^{226}Ra alpha decay. It is rather determined by the amount of ^{214}Pb and ^{214}Bi and the rate at which these two underwent beta and gamma decays. This can be further understood from the spectrum of the gamma before and after the heat-related drop.

A typical spectrum is given in Fig. 3, with some of the well known peaks of the ^{214}Pb – ^{214}Bi lines clearly recognizable. For example the ^{214}Pb (295, 351 keV), ^{214}Bi (609, 768, 934, 1120, 1377–1407, 1764, 2204, 2447 keV). ^{226}Ra /nano-Pd/ZrO₂. The spectra were obtained with two different spectrometers, the ^{125}RS and the Ortec. The former gives better resolution at lower energies while the latter is in general far more sensitive to give a better statistics. Overall, the spectra are very similar in the characteristic peaks.

It is also clear from the experiment that the characteristic shape of the spectrum does not change, at least in any discernable way to naked eyes, before and after the activity drop. We will see later in Fig. 6, the ^{214}Pb and ^{214}Bi activity may have a different time-dependent change profiles, and so in principle, there should exist small relative count change between the ^{214}Pb and ^{214}Bi peaks of Fig. 3 at different time. However this change is small and thus usually deeply buried beneath the fluctuation noise.

Because all peaks drop more or less evenly, the activity change can be at least explained by the change on the decay daughters like ^{214}Pb , ^{214}Bi . There are two possibilities: either the gamma decay rate changes or the amount of the ^{214}Pb and ^{214}Bi changes. While we cannot absolutely rule out the former, this is a difficult undertaking. Nuclear decay has different modes, which different susceptibility to the environment. It is rather difficult to affect the nuclear EM dipole transition rate with the chemical environment change when the gamma radiation is on the order of 1 MeV: The internal conversion mode, which is susceptible to orbital electrons, typically affects the keV gamma. Also, it is generally difficult to change the rate in the beta or alpha decay. Any possible gamma rate change here involves γ -rays of 100 keV and above and therefore is very unlikely. On the other hand, the concentration of the decay daughters can indeed change at relative ease.

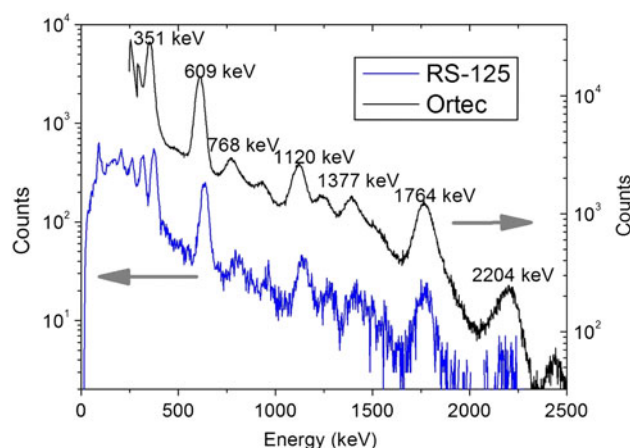


Fig. 3 Gamma spectrum of the experimental decay chain from the ^{226}Ra /nano-Pd/ZrO₂ powder

Here we give a real example. For many radiochemists, it is a well known fact that if a solution of radium nitrate is sealed tight in a brass container, the activity can go up with time in the course a few weeks/months until a saturation point is reached. The mechanism is actually straightforward to understand. With a chemically pure ^{226}Ra , the only radioactivity is from the alpha decay. The decay product ^{222}Rn is a gas and easily evolves and gets lost to the environment from the $\text{Ra}(\text{NO}_3)_2$ solution due to air convection when not kept in a gas-tight container. With a hermitically sealed container, the situation is different. The Rn gas cannot get out. Instead it further accumulates and decays, generating additional radioactivity, and transmutes into ^{214}Pb , then ^{214}Bi and so on. The daughters will then generate certain level of activity as well. In a steady state when an equilibrium is established, the decay chain then tends to have a total activity that is several times higher than the original activity of ^{226}Ra alone, due to the generation and presence of radioactive daughters.

From the aforementioned example, it is clear that there is a partition pathway where apparent radioactivity becomes susceptible to changes in the chemical environment if the decay chain involves a product capable of entry to the gas phase.

The very fact that the “hot-shot” tend to give the strongest positive results is yet further evidence that the drop apparent radioactivity is in large part due to radon loss or its diffusion out from the view-field of the spectrometer. The hot-shots were sealed and stored for a month and hence have an accumulation of radon, which gives higher activity compared to say a freshly prepared one. Then with the experiment starting and heating applied the radon gas may diffuse around and make the whole test cell of Fig. 1 a diffusive gamma source. For example, the radon gas may stray into the stainless steel tubes of Fig. 1 which is far from the effective “view-field” of the gamma detector. Then a drop in the activity that is counted by the spectrometer is a logical consequence. We cannot rule out the loss of radon to the gas line leakage as well, which should also give a drop in the gamma count.

Theoretical understanding of the results

If the diffusion/accumulation of the radon gas is the reason for the change of radioactivity during the heating and the later-on recover stage, we want to see if we can qualitatively reproduce the experimental phenomenology from the theory. In the rise and fall of the radioactivity count, there exists a peculiar asymmetry. The drop is relatively fast in a course of a few hours, but the count recovery to the pre-heating value is a long process of a few days or more. We will see in the following that this can be fully understood

within our current knowledge of the decay rates in the ^{226}Ra decay chain, as long as the Rn loss due to heating is properly taken into account.

The ^{226}Ra chain is given fully in Fig. 4. However, due to the fact that most of the gamma spectrum is from the ^{214}Pb and ^{214}Bi , we only need to stop the chain by ^{214}Bi . This also makes the decay chain free of branches, much simplifying our differential rate equations. The simplified chain is given in Fig. 5.

The decay dynamics is governed by a set of differential equations. First let us study the peculiar drop after the heat is turned on. The decay was in a steady state before the increase of the temperature to 125°C at $t = 0$. Assume that all radon is able to diffuse out of the gamma detector's field-of-view at $t = 0$ due to the heating. Of course this is oversimplification, but we see it will reproduce most of the observed phenomena. Let X , Y , Z be the amount of Po, Pb and Bi, all of which are a function of t and with the initial values at $t = 0$ given by X_0 , Y_0 , Z_0 . Because the chain is now broken at ^{222}Rn , we only need to consider X , Y , Z . The set of three simultaneous differential equations are:

$$\begin{aligned}\frac{dX(t)}{dt} &= -\frac{1}{a}X(t), & X(t=0) &= X_0 = \frac{a}{A} \\ \frac{dY(t)}{dt} &= \frac{1}{a}X(t) - \frac{1}{b}Y(t), & Y(t=0) &= Y_0 = \frac{b}{A}, \\ \frac{dZ(t)}{dt} &= \frac{1}{b}Y(t) - \frac{1}{c}Z(t), & Z(t=0) &= Z_0 = \frac{c}{A}\end{aligned}\quad (1)$$

where A is the decay half life of ^{226}Ra , which is 1,602 years. The half lives of ^{218}Po , ^{214}Pb and ^{214}Bi are given by a , b , and c , which are also shown in Fig. 5. The simultaneous equations above can be *rigorously* solved to give

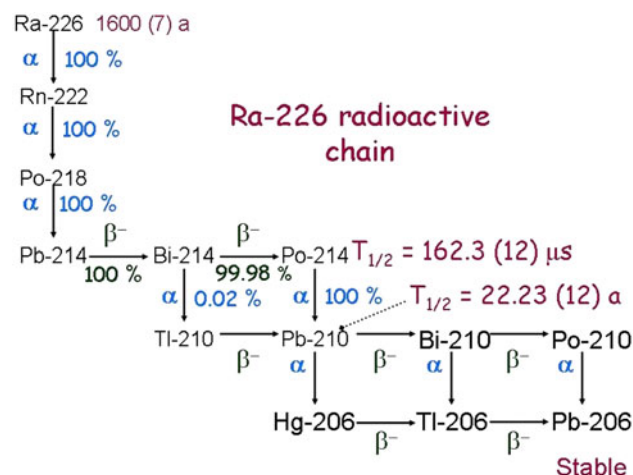


Fig. 4 The ^{226}Ra decay chain neglecting the very small branch of ^{218}At

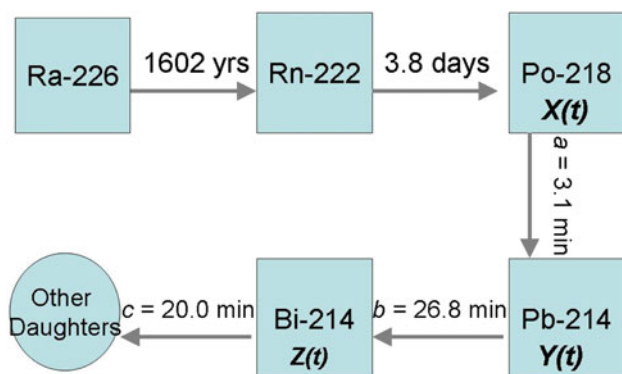


Fig. 5 The simplified ^{226}Ra decay chain

$$\begin{aligned}
 X(t) &= \frac{a}{A} e^{-t/a} \\
 Y(t) &= \frac{-b}{A(b-a)} (ae^{-t/a} - be^{-t/b}) \\
 Z(t) &= \frac{c}{A(b-c)(c-a)(c-a)} [(bc^2 - ac^2)e^{-t/c} \\
 &\quad + (ab^2 - cb^2)e^{-t/b} + (ca^2 - ba^2)e^{-t/a}]
 \end{aligned} \quad (2)$$

Plugging in the respective value of a , b , c and A , the relative change of ^{214}Pb with time can be shown as follows in Fig. 6 as the blue curve. The amount of ^{226}Ra is taken as 1.

Two features of Fig. 6 are notable. First, the characteristic time of the drop is on the order of ~ 45 min, longer than the 26.8 min intrinsic decay time of the ^{214}Pb itself. This drop is no longer a simple exponential decay. Second, as one can rigorously prove and have seen from Fig. 6 as well, the slope of the blue curve at $t = 0$ is actually zero. Therefore, even with an abrupt loss of radon, the drop in the gamma radioactivity count is not going to be immediate. Rather there is a short delay at the beginning, characterized by the zero slope at $t = 0$. Very similar features are seen in the time-dependent variation of the ^{214}Bi content, shown in Fig. 6 as the red curve. The differences with ^{214}Pb are two-fold. First, the characteristic decay time of

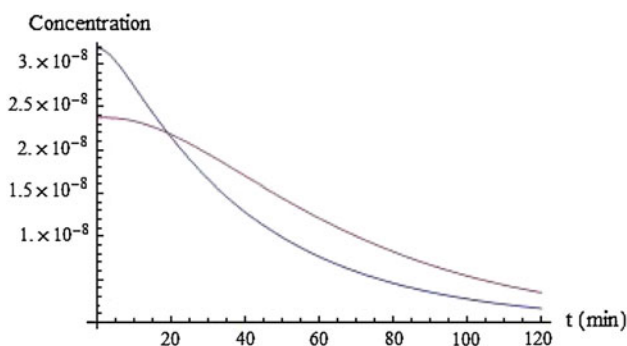


Fig. 6 The time dependence of the amount (concentration) of ^{214}Pb (blue) and ^{214}Bi (red) relative to ^{226}Ra

the ^{214}Bi is even longer, on the order of 80 min. Second, the initial delay at $t = 0$ in the drop is even longer: It may delay up to 20 min.

The combination activity of the Pb and Bi concentration of Fig. 6 then gives the time variation of the gamma counts, which is plotted in Fig. 7. The characteristics are fully borne out in the experiment (i.e., Fig. 2). Notice the prominent feature of initial delay in the activity drop. This makes the curve a concave type around $t = 0$. As time goes on and with the drop becoming more appreciable, the activity curve starts changing into a convex type, beyond a certain time frame.

The experimental count-versus-time curves (i.e., Fig. 2) have a characteristic drop time even longer than predicted by the theory (Fig. 7). This is fully understandable considering facts as follow. First, it is unlikely the heating can diffuse all of the radon. A significant portion is still left over and attached in the solid state lattice of the Ra-Pd powder. Second, the diffusion of the migrating radon is not an instant one. Rather it should take some time to diffuse. Taking the two factors into account, then one can readily understand the numerical differences between Figs. 2 and 7. Overall, the experimental drop time of a few hours is well understood from the decay rate equations and the practical considerations such as diffusion delay.

Now we switch our discussion to the recovery time. The relevant physics process is naturally assumed as follows. With the heating stopped the temperature T drops to a certain range when the diffusion/loss of Rn becomes negligible. We take this time as $t = 0$. Then we start to observe the accumulation of Rn over time. In this problem, we basically only have to deal with the first two legs of the decay chain in Fig. 5. Two simultaneous equations can be written in a fashion similar to Eq. 1 and then rigorously solved. Only the result is quoted here:

$$\text{Rn}(t) = \frac{\tau_2}{\tau_1 - \tau_2} (e^{-t/\tau_1} - e^{-t/\tau_2}), \quad (3)$$

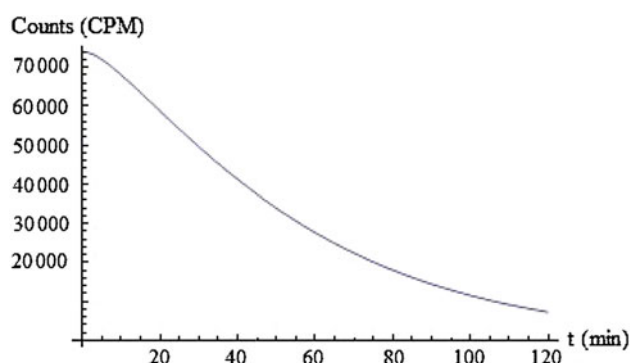


Fig. 7 Theoretical time dependence of the gamma activity drop

where $Rn(t)$ is the amount of radon versus time, with 1 as the amount of radium. τ_1 is 1602 years, which is $\gg \tau_2$, or 3.8 days. Under this condition,

$$Rn(t) = \frac{\tau_2}{\tau_1} (1 - e^{-t/\tau_2}). \quad (4)$$

The ^{222}Rn fraction can then be plotted graphically in Fig. 8. Note that the characteristic accumulation time is the order of ~ 5 days, which is actually from the radon half life of 3.8 days.

The predicted radon recovery from theory in Fig. 8 then can very well reproduce the count recovery experiment results of Fig. 9. We notice that there is slight a delay of radon accumulation and manifestation in the activity count. This is understandable from the following fact: there is a delay of a few days from radon to gamma radiation from $^{214}\text{Pb}/^{214}\text{Bi}$ via ^{218}Po . The close resemblance of Figs. 8 and 9 is yet another strong indication that we are on the right track to understanding the phenomenon.

The qualitative and quantitative agreements between the experimental results and our theoretical model are satisfactory at this stage considering some important unknowns such as radon diffusion rate and its desorption kinetics in a nanostructured material. Note that so far we did not utilize any assumption in the change of the intrinsic nuclear decay rate. Also keep in mind the difference between the intrinsic decay rate and the observed (apparent) radioactivity count. The lesson that we learned from this investigation is two-fold. First, even if the intrinsic decay rate does not change with chemical environment, in an actual experimental a radioactivity count may still change with the environment. In fact, it may change a lot, depending on the counting methods, loss of daughter gases, leaching of liquids and/or the time accumulation of the decay daughters. Second, an observed change in the radioactivity count does not necessarily mean a change in the intrinsic decay rate, although it generally does not rule out that the decay rate change either.

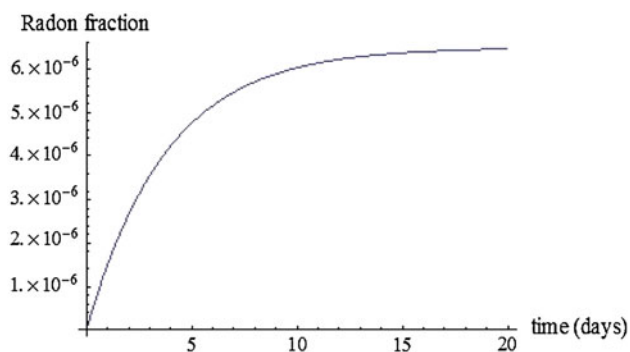


Fig. 8 The theoretical time dependence of radon accumulation after the heating is stopped at $t = 0$

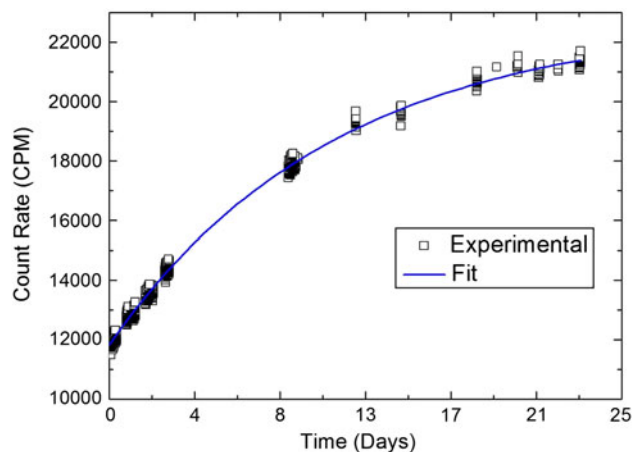


Fig. 9 The experimental time dependence of the activity count after heating is stopped. Squares are the experimental result for the test with deuterium. The blue line is the fit using Eq. 4 and Fig. 8

Possible temperature change in the detector

Although the radioactivity probes are in general 2 cm away from the sample, the scintillation crystals themselves are typically covered by some protective covers and therefore it is difficult to directly measure crystals' temperature and its possible change. Therefore, a blank experiment was designed to test any possible temperature effect on the detector (Fig. 10).

A blank radioactive cell of ^{226}Ra is fabricated so that any possible escape of the radon gas and other volatile decay daughters is eradicated by design. Compared to the ordinary capsule design of Fig. 1, there are two major differences: (1) the stainless tubes are replaced by solid stainless wires so that there is not a flow of gas during the heating. (2) The ^{226}Ra loaded powder and its radioactive decay products are hermitically sealed in the Vycor capsule by flame-fusing the Vycor to stainless steel wires. As the

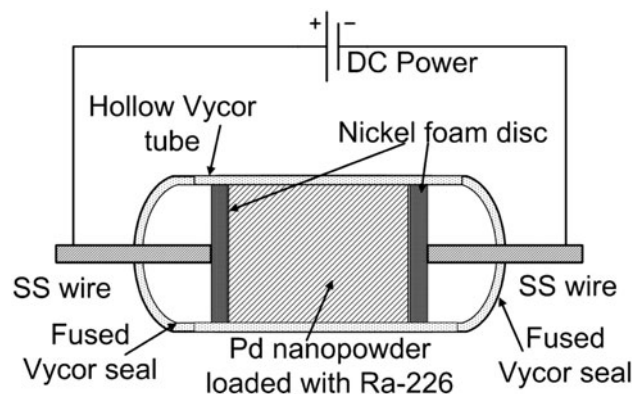


Fig. 10 The detail of the blank test capsule. The seals on the two ends are by flame fusing the Vycor tubes to the steel wires, instead of the Devcon 5-min epoxy. This gives almost sure proof against the leaking of radon

result the volatile radioactive species are always kept inside. This blank capsule, once flame sealed, is kept for over 1 month so that its activity nearly reaches the equilibrium level.

Then the blank capsule is heated with electric current to a temperature up to 130 °C. Of all the properly sealed blank cells, no appreciable change of radioactivity count (>2%) was ever recorded. This observation rules out temperature as the possible cause of significant variations in the radioactivity counting.

Implication to decay rate experiments

It is now clear from our experiment that the migration of the isotopes in the radioactive decay chain may pose a serious problem if the isotopes are allowed to migrate in the counting process. The effects come in very different sometimes unexpected ways. Take the radioactive counting of ^{22}Na and ^{198}Au for example. ^{22}Na positron-decays into ^{22}Ne which is a gas and gives a 1274.6 keV gamma. The gaseous ^{22}Ne definitely gets lost if not kept in a gas tight environment at the room temperature, and this may cause spurious effect. ^{198}Au beta-decays into ^{198}Hg which is volatile at room temperature. The counting at room temperature and a cryogenic one then risks having different consequences. The counting of ^{198}Au beta activity is often through the 412 keV gamma, which is actually a property of the excited ^{198}Hg nuclei. The room temperature counting of ^{198}Au is basically without the high-Z daughter product to absorb the gamma rays, and therefore should give larger count, which may be attributed to a higher decay rate at room temperature. Of course exactly how the volatile daughter or their absence might affect the counting depends on the detail of sample condition and geometry. So it is not easy to draw a conclusion as to whether it played a role or not in some of the previous discussed experiments. However a precaution with this in mind will definitely help future experiments to reach better accuracy.

Sometimes the property of detector may play a role. In the current study, if the spectrometer has a larger sensitive field, the gamma daughters from the astray radon would still have shown up in the count and so the drop would not as dramatic. Similar detector effect might have played a role in the reported drop of tritium in Ti [16]. Note that a not-so-sensitive Geiger counter is utilized in the counting of the X-rays from bremsstrahlung of betas from the tritium decay. The initial drop might due to that external bremsstrahlung of the tritium was reduced as it desorbs from the Ti metal matrix. The beta from the tritium is generally low in energy, 18.6 keV maximal, and in fact only 5.7 keV in average. The bremsstrahlung thus generated is only 1 keV or less. X-ray of this energy can only penetrate a few

micrometers' titanium. Therefore, only the tritium a few micrometers with the Ti surface were actually counted. As the desorption goes on, the surface is first depleted of the tritium which then contribute the initial loss. Of course, this aforementioned scenario does not rule out the author's explanation so far. However, in many cases, a correct account of the observed activity change involves a lot of disciplines even within the field of physics itself.

Sometimes the change in the radioactivity count can come in a very surprising way. One example is given by surface segregation, as suggested in [17]. In this case, ^{210}Po is found to undergo significant migration in lead and this affects the apparent count of an alpha counter. More precisely, the migration is due to a surface segregation of the ^{214}Po in lead. What is surprising is that the segregation/diffusion of ^{210}Po can happen at a very low temperature of 50–100 °C, and this involves a diffusion of quite large atoms such as ^{210}Po , whose size would otherwise give us an impression of negligible diffusion at 100 °C.

The current experiment does not completely rule out an intrinsic change of the decay rate with temperature or the heating current. To achieve this, a more quantitative study of the variation in the activity count as a function of temperature should be carried out. Or alternatively, an experiment in a decay chain without gaseous daughters is needed, such as the ^{60}Co . This is a study that is currently under our investigation with results to be published later. The diffusion coefficient of radon in the lattice should follow an Arrhenius law. Therefore, the rate by which the activity count changes in a heating experiment depends on temperature. The question as to whether Rn diffusion is the sole reason for the activity change hence hinges upon verification of the Arrhenius law. This should constitute an ideal target for further studies.

Discussion and conclusion

We carried out a study of radioactivity variation during the heating of radium supported on the nanopowders of palladium. The studies were carried out with the help of a variety of gases during the experiments. It is found that flushing hydrogen isotopes typically gives the most prominent change. This is understood though not proved with direct evidences that hydride forming in both Ra and Pd helps expanding the metal lattice thus facilitating the release of the Rn.

An attempt to explain the change in terms of Rn loss was given and found to give satisfactory results compared to the experiment. We proposed that similar conditions in various experiments on decay rates may pose complication to an accurate determination of the decay rate. This study also helps to shed light to some decay rate experiments

where the complication may cause erroneous interpretation of results.

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