22.01 Fall 2016, Problem Set 3 Solutions

October 9, 2016

Complete all the assigned problems, and do make sure to show your intermediate work.

---

1 Activity and Half Lives

1. Given the half lives and modern-day abundances of the three natural isotopes of uranium, calculate the isotopic fractions of uranium when the Earth first formed 4.5 billion years ago.

   Today, uranium consists of 0.72% ^{235}U, 99.2745% ^{238}U, and 0.0055% ^{234}U. *However*, it is clear that the half life of ^{234}U (245,500 years) is so short compared to the lifetime of the Earth (4,500,000,000 years) that it would have all decayed away had there been some during the birth of the Earth. Therefore, we look a little closer, and find that ^{234}U is an indirect decay product of ^{238}U, by tracing it back from its parent nuclides on the KAERI table:

$$^{238}U \xrightarrow{\alpha} {}^{234}Th \xrightarrow{\beta^-} {}^{234}Pa \xrightarrow{\beta^-} {}^{234}U \tag{1}$$
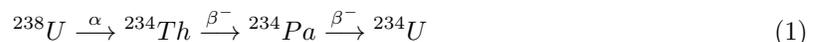
   Therefore we won't consider there being any more ^{234}U than would normally be in equilibrium with the ^{238}U around at the time. We set up the two remaining equations as follows:

$$N_{235} = N_{0_{235}} e^{-t/t_{1/2},235} \qquad N_{238} = N_{0_{238}} e^{-t/t_{1/2},238} \tag{2}$$

   Using the current isotopic abundances from above as N_{235} and N_{238}, the half lives from the KAERI Table of Nuclides $\left(t_{1/2,235} = 703800000\,y;\ t_{1/2,238} = 4.468 \cdot 10^9\,y\right)$, and the lifetime of the earth in years (keeping everything in the same units), we arrive at the following expressions for $N_{0_{235}}$ and $N_{0_{238}}$:

$$N_{0_{235}} = \frac{N_{235}}{e^{-t/t_{1/2},235}} = \frac{0.0072}{e^{-4.5\cdot10^9/7.038\cdot10^8}} = 4.307 \qquad N_{0_{238}} = \frac{N_{238}}{e^{-t/t_{1/2},238}} = \frac{0.992745}{e^{-4.5\cdot10^9/4.468\cdot10^9}} = 2.718 \tag{3}$$

   Finally, taking the ratios of these two relative abundances gives us absolute abundances:

$$f_{235} = \frac{4.307}{4.307 + 2.718} = 0.613 \qquad f_{238} = \frac{2.718}{4.307 + 2.718} = 0.387 \tag{4}$$

   ^{235}U was 61.3% abundant, and ^{238}U was 38.7% abundant. Imagine how much easier it would have been to make nuclear reactors during the Pre-Cambrian period!

2. Explain the principle behind radioisotope carbon dating. Look up the Shroud of Turin, the supposed burial cloth of Jesus of Nazareth. What would be the isotopic fraction of ^{14}C expected if the Shroud of Turin was real? What was the actual isotopic fraction of ^{14}C, and how old does that make it?

   Radioisotope carbon dating looks at the amount of ^{14}C in a material, assuming that while it was alive (either a living organism like an animal, or fibers taken from a living plant/tree, etc.) it was in equilibrium with the normal isotopic fraction of ^{14}C in the

environment, which is roughly one part per trillion (ppt). This fantastically small, but very regular, amount of ^{14}C is produced from proton capture by cosmic-ray induced neutron capture from ^{14}N, the most abundant isotope in the Earth's atmosphere:

$$^{14}_{7}N +^{1}_{0} n \longrightarrow^{14}_{6} C +^{1}_{1} p^{+} \tag{5}$$

This ^{14}C remains in equilibrium with all living things in the carbon cycle. Therefore, by counting the number of natural β^- decays coming out a carboniferous material, one can determine the percentage of ^{14}C inside, and therefore the approximate age of the specimen.

Such is the case for the Shroud of Turin, supposedly the burial cloth of Jesus of Nazareth. If it were truly so, then it should be 2,000 years old, and the fraction of ^{14}C remaining should have been:

$$N_{14_C} = N_{0_{14_C}} e^{-2,000/5,760} = (1\, ppt)\,(0.056) = 0.707\, ppt \tag{6}$$

which should yield 0.0225 β^- particles per second per gram of material. This does not account for abnormal rates of production of ^{14}C, such as from gigantic solar flares, supernovas, or nuclear weapons testing.

Three labs independently tested swatches of the cloth along with three control specimens, and found the average age to correspond to 691 years old, which would have yielded an actual isotopic fraction of 0.887 ppt.

2   RTG Operation

In these problems, consider the decay of ^{239}Pu, the isotope used in radioisotope thermoelectric generators (RTGs). (**NOTE: This was a typo, ^{238}Pu is actually used for RTGs. To maintain consistency with the problem statement, we will solve the problems for ^{239}Pu.**)

1. Write the two possible types of decay reactions for ^{239}Pu, and state which decay processes (and competing processes) may be possible for each general type of reaction. You don't have to address every single energy level, there are dozens! Just group them into categories.
   Two possible decay chains are possible for ^{239}Pu, an alpha decay with subsequent gamma decays (ITs) and spontaneous fission:

$$^{239}Pu \rightarrow^{235} U + \alpha + m\gamma; \qquad m = 0 \dots lots \tag{7}$$

$$^{239}Pu \rightarrow FP_1 + FP_2 + 2^1_0 n \tag{8}$$
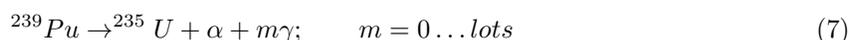
No processes compete with alpha decay, though each IT (gamma decay) competes with internal conversion (IC). When IC occurs, the gamma ray may instead eject an electron with energy $E_\gamma - E_{binding}$, followed by either photon emission from a higher shell electron falling back down to this newly opened lower level, or an Auger electron. Nothing directly competes with spontaneous fission, though the fission products (FPs) can undergo any number of their own decays, ranging from beta to positron/electron capture, to neutron emission, to gamma/IT, to alpha decay themselves. Many fission products undergo beta decay.

2. Now consider only the *three* most likely alpha decay energies of ^{239}Pu.

   (a) Draw a complete energy level diagram showing alpha decay to these three energy levels, and any possible, successive decays to the ground state.
      The three most likely alpha decays can be found by consulting the KAERI table of nuclides at http://atom.kaeri.re.kr:8080/cgi-bin/decay?Pu-239%20A. In addition, thinking ahead, the following electron level binding energies and transitions were found on the NIST X-ray transition energies database at:

. The "K-edge" energy corresponds to the binding energy of a K-shell electron, while the K1L1 transition corresponds to a transition from the most bound L-level to the most bound K-level:. Other numbered transitions were not chosen, as they are very close to the 1-1 transitions, and we didn't worry about the N-shells:

Energy (keV)	Intensity (rel.)	Transition	Energy (keV)
5244.4	0.03	**K edge**	**121.8**
5156.6	**73.3**	**L edge**	**23.1**
5144.3	**15.1**	**K-L**	**98.7**
5111.2	0.03	**K-M**	**115.9**
5105.5	**11.5**	**L-M**	**17.2**

Remember that the energy of the alpha decay is not the same as the Q-value, because the ^{235}U recoil nucleus also takes away some of that kinetic energy released by the Q-value. One can use conservation of momentum to find out the corresponding Q-value for each alpha energy:

$$p_\alpha = p_U \tag{9}$$

$$m_\alpha v_a = m_U v_U \tag{10}$$

$$\sqrt{2m_\alpha E_\alpha} = \sqrt{2m_U E_U} \tag{11}$$

$$E_U = \frac{m_\alpha}{m_U} E_\alpha \tag{12}$$

We know the energy of the alpha particle in each case, and using this formula, we can find the corresponding Q-value for each alpha energy:

$$Q = E_\alpha + E_U = E_\alpha + \frac{m_\alpha}{m_U} E_\alpha = E_\alpha \left(1 + \frac{m_\alpha}{m_U}\right) = E_\alpha \left(1 + \frac{4.0026032\,amu}{235.0439231\,amu}\right) \approx 1.017029 E_\alpha \tag{13}$$

This yields the following table of Q-values for our three alpha particle energies, and the corresponding ^{235}U excited states by subtracting these Q-values from the ^{241}Am relative ground state energy (5244.5 keV):

E_α (keV)	Q (keV)	^{235}U Energy Level (Calc.)	Closest ^{235}U Level (KAERI Table)
5156.6	5244.4	0.1	0.1
5144.3	5231.9	12.6	13.0
5105.5	5192.4	52.1	51.7

Clearly the values are remarkably close. The full decay diagram proceeds as follows:

One can see that not every energy transition is allowed. The following transitions were identified, along with their corresponding gamma ray energies. In addition, allowed electron transitions following internal conversion are tabulated for just the first four energy levels:

E_i	E_f	ΔE	K-eject?	L-eject?
51.7	0	51.7		✓
51.7	13.0	38.7		✓
46.2	0	46.2		✓
13.0	0	13.0		

Here, there are no K-ejections allowed for internal conversion *for just the most likely three alpha decay energies*. Higher energy states can allow this to happen, just not in our simplified diagram.

(b) For each of the initial alpha particle energies, separately sketch a hypothetical photon (gamma plus x-ray) spectrum that you would expect to observe. You may want to use the NIST X-Ray Transition Energy Database to help generate your answer.

This spectrum should consist of all the gamma ray energy differences in the table above, along with all the L-level x-ray transition energies identified, as no IT/IC process is energetic enough to eject a K-shell electron.

3. It is clear that ^{239}Pu produces a few types of radiation at many different energies. Do you expect the alpha particles, the gamma rays, the spontaneous fissions, or the x-rays to be responsible for producing the most heat generation in an RTG, and why?

The alpha particles should be responsible for most of the heat, because they interact so much more strongly with matter, and therefore the electron clouds in the matter. Their heavy mass and high charge compared to gammas or x-rays (no mass, no charge) gives them far higher ionizing power, and therefore they will all stop within the RTG to deposit their kinetic energy as heat. Spontaneous fission is so rare that while it does produce more energy per disintegration than any other method, it just doesn't happen often enough to matter.

3 Medical Isotope Physics

In these problems, consider the decay of ^{99}Mo, a crucial medical isotope widely used in imaging and diagnosis procedures.

1. Calculate the Q-value for the decay of ^{99}Mo using the binding energies of the initial and final nuclei, and any other information that you need.

The decay of ^{99}Mo proceeds by beta decay to ^{99m}Tc, followed by a gamma decay (IT) to stable ^{99}Tc:

$$^{99}Mo \to\,^{99m}Tc + \beta^- + \bar{\nu}_e\,^{99m}Tc \to\,^{99}Tc + \gamma \tag{14}$$
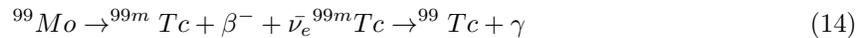
All that is required are the masses in amu of ^{99}Mo and ^{99}Tc, and the conversion factor between amu and MeV:

$$Q = \left(m_{Mo-99}\cancel{c^2} - m_{Tc-99}\cancel{c^2}\right)\left(\frac{931.49\,MeV}{amu - \cancel{c^2}}\right) = 931.49\,(98.9077116 - 98.9062546) = 1.3572\,MeV \tag{15}$$

This is indeed the difference in energy levels according to the decay diagram for Mo-99:
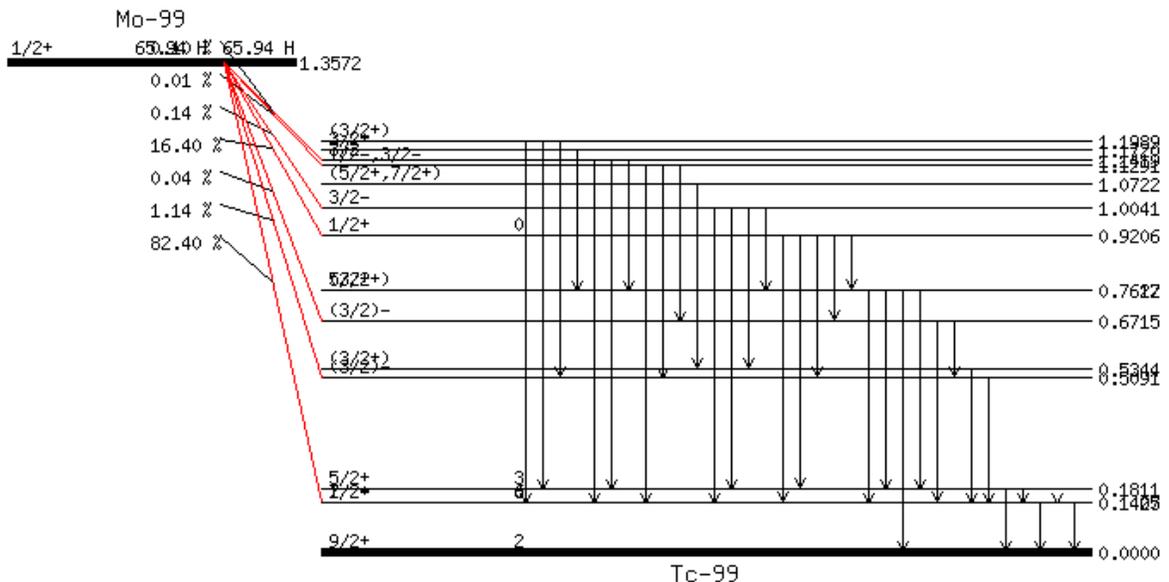
Courtesy of Korea Atomic Energy Research Institute. Used with permission.

4

2. You may have noticed that ^{99}Mo is an unstable isotope. Which nuclear reactions could create ^{99}Mo? Write the nuclear reactions for these processes, and calculate their Q-values to justify your answer. **This is where some creativity can come into play.** ^{99}Mo **could either be produced by spontaneous decay reactions, or by deliberate bombardment of parent isotopes with other particles. First, the decay modes:**

$$^{103}Ru \to^{99} Mo + \alpha; \qquad Q = (m_{Ru-103} - m_{Mo-99} - m_\alpha)\cancel{c^2}\left(\frac{931.49\,MeV}{amu - \cancel{c^2}}\right) = -3.71\,MeV \qquad (16)$$

$$^{99}Nb \to^{99} Mo + \beta^- + \bar\nu; \qquad Q = 1.3572\,MeV\ (above) \qquad (17)$$

$$^{99}Tc \to^{99} Mo + \beta^+ + \nu; \qquad Q = -1.3572\,MeV \qquad (18)$$

$$^{99}Tc \to^{99} Mo + e^-\ (EC); \qquad Q = -1.3572\,MeV \qquad (19)$$

$$^{100}Mo \to^{99} Mo + n; \qquad Q = -8.29\,MeV \qquad (20)$$

$$^{235}U \to^{99} Mo +^{134} Sn + 2^1_0n; \qquad Q = 177.4\,MeV \qquad (21)$$

Now for the energetic particle bombardment methods:

$$^{98}Nb + p^+ \to^{99} Mo; \qquad Q = 9.73\,MeV \qquad (22)$$

$$^{98}Mo + n \to^{99} Mo; \qquad Q = 5.93\,MeV \qquad (23)$$

$$^{95}Zr + \alpha \to^{99} Mo; \qquad Q = 2.73\,MeV \qquad (24)$$

As one can see, only beta decay and spontaneous fission is possible, though a number of bombardment options are available.

4   Allowable Nuclear Reactions

For these problems, determine whether the following reactions would be allowed, and answer the additional questions.

1. Which of the following decay methods are energetically allowable from the ground state of ^{216}At? Back up your reasoning with an energetic argument.

   (a) Alpha decay
   $$^{216}\text{At} \to^{212} Bi + \alpha; \qquad Q = 7.949\,MeV \qquad (25)$$

   Allowed, plus we get exactly the total alpha decay energy in the KAERI table.

   (b) Beta decay
   $$^{216}\text{At} \to^{216} Rn + \beta^- + \bar\nu; \qquad Q = 2.003\,MeV \qquad (26)$$

   Allowed, plus we get exactly the total beta decay energy in the KAERI table.

   (c) Positron decay
   $$^{216}\text{At} \to^{216} Po + \beta^+ + \nu; \qquad Q = 0.469\,MeV \qquad (27)$$

   Not allowed, because the Q-value isn't large enough to create the positron (1.022 MeV).

   (d) Electron capture
   $$^{216}\text{At} \to^{216} Po + \nu; \qquad Q = 0.469\,MeV \qquad (28)$$

   Allowed, plus we get exactly the total electron capture decay energy in the KAERI table.

(e) Isomeric transition

$$^{216}\text{At} \rightarrow ^{216}At; \qquad Q = 0 \tag{29}$$

Impossible, because ^{216}At is already at its ground state.

(f) Spontaneous fission

    i. Can you find an instance where this particular one is energetically allowable?
    Many of them are, here is one example:

$$^{216}\text{At} \rightarrow ^{114}_{45} Rh + ^{100}_{40} Zr + 2^1_0 n; \qquad Q = 138.3\, MeV \tag{30}$$

    ii. Why do you think it's never observed?
    Just because a reaction is energetically allowable does not mean that it will happen. In order for a nucleus to spontaneously fizz, it must overcome a significant strong nuclear force attractive barrier. Think of it as a huge activation energy, required to release a huge amount of energy at the end of the reaction. The higher this barrier, the less likely a particular nuclear vibration will allow for the fission products to separate themselves from the nucleus. These barriers are typically on the order of 200 MeV, which is why only the super-heavy elements undergo spontaneous fission with any measurable probability.

2. For the reactions which are allowed, write the full nuclear reaction in each case, and draw a graph of the energy spectrum you would expect to see from each released form of radiation, including secondary ejections of particles or photons.
**See above for the full nuclear reactions.**

For alpha decay, one would expect to see mono-energetic alpha particles at the following tabulated energies: 7802 keV, 7683 keV, 7610 keV, 7560 keV, 7470 keV, 7390 keV, 7317 keV, 7240 keV. The diagram does not state any allowable gamma decays, though in reality they are likely to be observed. The short half-life (0.3 ms) of ^{216}At is likely to blame for this lack of measured data. Therefore, without knowing the selection rules for gamma emission from spin and parity states (to be learned in 22.02), all that one can say is that the maximum energy available to eject an electron is that of the largest possible transition (573 keV). Using the NIST x-ray transition energy tables for ^{216}At, the highest energy gamma ray could only K-shell or L-shell electrons (binding energies of 95.7 keV and 17.5 keV, respectively). Therefore, other photon transitions of KL, KM, KN, LM, or LN transitions may be observed from IC processes competing with gamma emission. One would also expect to observe Auger electrons at these energies, in addition to the K, L, M, N... conversion electrons. See the diagrams below:
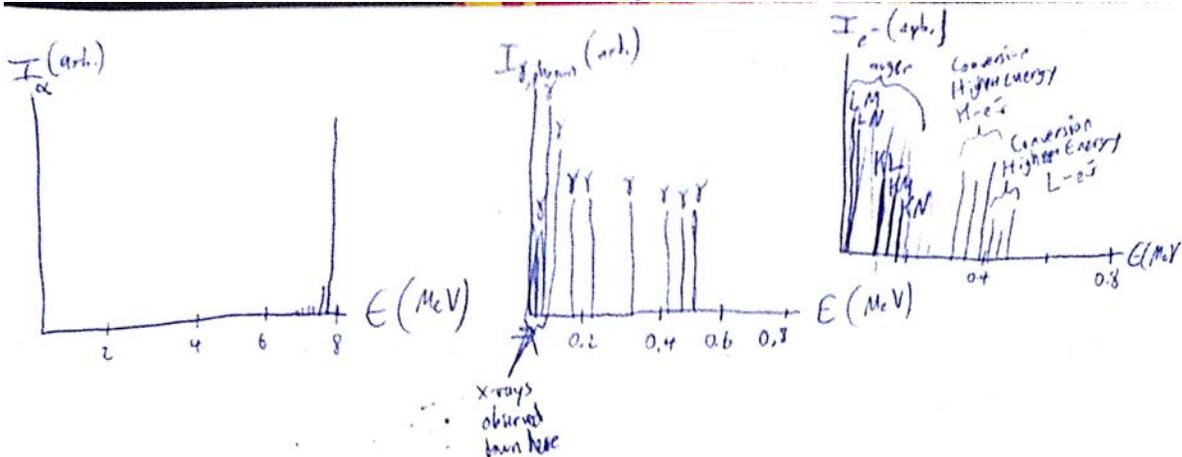
For beta decay, one would expect to see an electron spectrum as below, with E_{max} at the

Q-value of 2.003 MeV. The anti-neutrino spectrum must be its mirror image, because the probability of observing a beta with energy E is equal to observing an anti-neutrino with (Q-E).
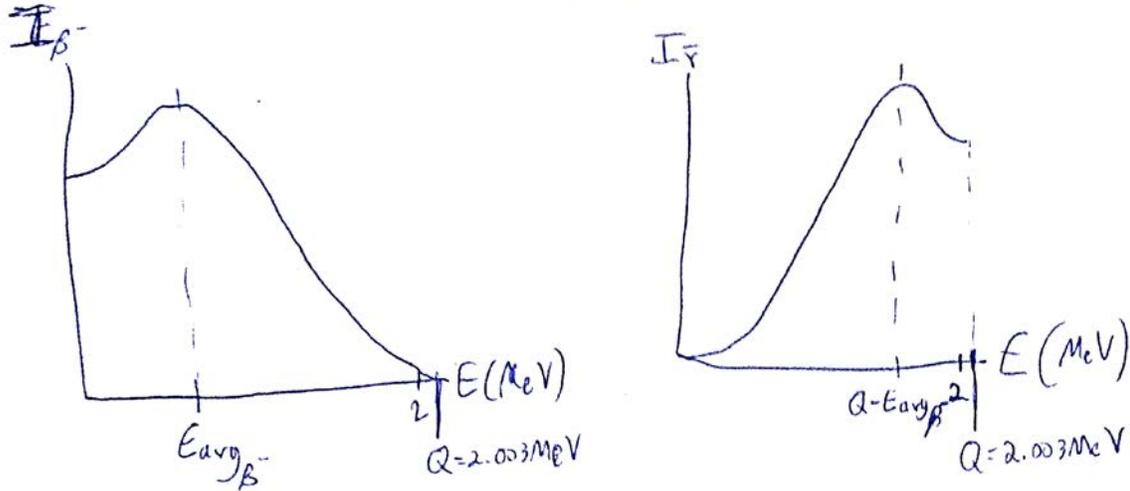
For electron capture, one would only expect to see the transition x-rays and corresponding Auger electrons as seen in alpha decay, maxing out at 469 keV.

For spontaneous fission, one would expect to see a huge range of photons, conversion electrons, transition x-rays, and whatever decay products are possible from the fission products. Then again, even though it's energetically allowable, it's improbable.

3. For the reactions which are not allowed, under which conditions could they be allowable? In other words, how would you insert energy into the system to make them allowed, and how much?

   For all reactions that are not allowed, increasing the kinetic energy of the ^{216}At **nucleus to equal -Q (most reactions) or such that $Q + E_{At} \geq 1.022$ MeV (positron) would make this allowable. This is because a** *necessary and sufficient* **condition for a nuclear reaction to proceed is that the sum of Q and the kinetic energy are greater than zero:**

$$Q + E_i \geq \begin{cases} 0 & (most\ reactions) \\ 1.022\ MeV & (positron\ emission) \end{cases} \tag{31}$$

22.01 Introduction to Nuclear Engineering and Ionizing Radiation
Fall 2016