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U-235 and U-238 isotopic fractions of natural Uranium throughout geological time[1]

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Abstract: Throughout geological time natural Uranium has been composed to within a fraction of a percent of only two isotopes, U-235 and U-238. In this note, the abundance fractions of U-235 and U-238 and the ratio of U-235 to U-238 are computed throughout the earth's history. This computation is of interest since the U-235 isotope, which sustains nuclear reactions unlike the more copious U-238, was significantly more abundant in the past and induced natural operating reactors long ago in the earth.

Long-lived Uranium Isotopes in the Earth

The earth was formed from coalesced debris ejected from exploding supernovae, collisions of neutron stars, and other violent early universe events. The debris had significant amounts of heavy metals in it, including Uranium. In the processes that formed heavy metals, there was a more equal percentage of Uranium-235 and Uranium-238 than there is today. However, once the Uranium escapes the high pressure chemical forging environment of dense stars there is no more production of U-235 and U-238 and the passive activities of radioactive decay subsequently determine the changes of their abundances over time. Thus, left to their own devices, the U-235 becomes much less abundant than U-238 over time due to its much shorter mean lifetime.

In this note we compute the Uranium abundances going back in geological time. For this calculation we assume that a chunk of natural Uranium is entirely composed of U-235 and U-238. That is not quite right because there are trace amounts of other isotopes of Uranium, but those other isotopes have much shorter lifetimes and are less relevant as long as we are more than a few hundred million years past nature's violent era of heavy-element manufacturing, which is the case here.

U-235 is the isotope that enables sustained chain nuclear reactions, and so the higher the U-235 fraction of a sample the better it can be used toward nuclear energy or even nuclear bombs. Nuclear energy typically requires a few percent U-235 fraction, whereas nuclear bombs typically use over 90 percent enriched U-235 but need at least about 20 percent for straightforward construction (Brown & Glaser 2016), which is the threshold above which one designates the sample "highly enriched uranium" (HEU).

One of the reasons for the calculation of natural U-235 fraction in geological time is to understand when natural reactors were able to sustain nuclear reactions. The most famous example of this is the Oklo reactor in what is today Gabon, Africa. This natural Uranium fission reactor began sustained reactions about 1.7 million years ago (Myr ago) and lasted for a few hundred thousand years. The reactor was made possible by ground water that acted

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as neutron moderators and a copious supply of enriched Uranium (between 3 and 4 percent U-235 composition, compared to today's 0.7 percent).

Data and variable definitions

Before embarking on the calculations and the results let us review the relevant data needed.

The half-lives of U-235 and U-238 will be designated as τ_5 and τ_8 respectively. Their numerical values are (NNDC 2018)

$$\tau_5 = 0.704(1)\,\mathrm{Gyr}, \quad \text{and} \quad \tau_8 = 4.468(6)\,\mathrm{Gyr} \tag{1}$$

where Gyr means "giga years", or 10^9 years.

The half-life τ is related to the average lifetime T and the decay width Γ according to

$$\tau = \frac{\ln 2}{\Gamma} = T \ln 2, \quad \text{or equivalently} \quad \Gamma = \frac{1}{T} = \frac{\ln 2}{\tau}. \tag{2}$$

Similar to τ_5 and τ_8 we define Γ_5 and Γ_8 as the total decay widths of U-235 and U-238, and we define T_5 and T_8 as the mean lifetime of U-235 and U-238, respectively.

It will become useful in the next section to define the variable T_Δ, which is

$$T_\Delta = \frac{1}{\Gamma_5 - \Gamma_8} = \frac{1}{\ln 2}\left(\frac{1}{\tau_5} - \frac{1}{\tau_8}\right)^{-1} = 1.206\,\mathrm{Gyr}. \tag{3}$$

The fractions of U-235 and U-238 in a natural sample of Uranium today are designated as $\hat{f}_5$ and $\hat{f}_8$, respectively. Their numerical values are (NNDC 2018)

$$\hat{f}_5 = 0.007204(6), \quad \text{and} \quad \hat{f}_8 = 0.992742(10). \tag{4}$$

The isotopic fractions change over time and we can designate that change as $f_i(t)$ where $f_i(\text{today}) = \hat{f}_i$. These fractions as a function of time will be computed in the next section. The ratio of isotopic fractions is also time dependent and is designated as

$$\gamma(t) = \frac{f_8(t)}{f_5(t)}, \quad \text{where } \hat{\gamma} = \frac{\hat{f}_8}{\hat{f}_5} = 137.8. \tag{5}$$

Uranium isotopic fractions throughout geological time

The decays of an Uranium isotope over time follow the exponential law of radioactive decays:

$$N_i(t) = N_i(t_0)e^{-\Gamma_i(t-t_0)}. \tag{6}$$

Normally we are interested in investigating how much material is left after some time into the future. However, for us we are interested in how much material there was in the past given how much there is today. For that reason, we introduce a new variable $\tilde{t}$ which is "time ago." So $\tilde{t} = 0$ is today, and $\tilde{t} = 1.3\,\mathrm{Gyr}$ is 1.3 Gyr in the past.

With this new variable, and recognizing that U-238 does not decay into U-235 and vice versa, we can rewrite the radioactive decay law as

$$N(\tilde{t}) = \hat{N}_i e^{\Gamma \tilde{t}} \tag{7}$$

where $\hat{N}_i$ is the number of isotopes of type i that exist today.

We are not interested in keeping track of all the Uranium isotopes on the planet, and so wish to recast our equations in terms of fraction $f_i(\tilde{t})$ of an isotope within a sample found at some time $\tilde{t}$. The computation of this results from the definition of $f_i(\tilde{t})$. First, the result for $f_5(\tilde{t})$ is

$$f_5(\tilde{t}) = \frac{N_5(\tilde{t})}{N_5(\tilde{t}) + N_8(\tilde{t})} = \left(1 + \hat{\gamma}\, e^{-\tilde{t}/T_\Delta}\right)^{-1}, \tag{8}$$

and the result for $f_8(\tilde{t})$ is

$$f_8(\tilde{t}) = \frac{N_8(\tilde{t})}{N_5(\tilde{t}) + N_8(\tilde{t})} = \left(1 + \hat{\gamma}^{-1}\, e^{\tilde{t}/T_\Delta}\right)^{-1}, \tag{9}$$

where $\hat{\gamma}$ and T_Δ are given in the previous section. One can readily check that $f_5(\tilde{t}) + f_8(\tilde{t}) \equiv 1$ for all $\tilde{t}$ by construction from eqs. 8 and 9, which is an excellent approximation as discussed above.

There are two variables of interest in the early geological history. The first is the U-235 percentage factor, which is just the percent value representation of $f_5(\tilde{t})$ (see eq. 8). This percent abundance of U-235 as a function of $\tilde{t}$ ($100 f_5(\tilde{t})$) is plotted in Fig. 1.

The second variable of interest is the ratio of U-235 to U-238 multiplied by 100, which we can call $R_5(\tilde{t})$. It's the variable used by Gauthier-Lafaye and Weber (2003) to track U-235 abundance in early geological time. The computation of it yields :

$$R_5(\tilde{t}) \equiv 100 \left(\frac{f_5(\tilde{t})}{f_8(\tilde{t})}\right) = 100 \left(\frac{1 + \hat{\gamma}^{-1}\, e^{\tilde{t}/T_\Delta}}{1 + \hat{\gamma}\, e^{-\tilde{t}/T_\Delta}}\right). \tag{10}$$

A plot of R_5 vs. $\tilde{t}$ is presented in Fig. 2. The result matches that of Fig. 8 of Gauthier-Lafaye and Weber (2003).

References

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Gauthier-Lafaye, F., Weber, F. (2003). "Natural nuclear fission reactors: time constraints for occurrence, and their relation to uranium and manganese deposits and to the evolution of the atmosphere," *Precambrian Research* 120, 81.

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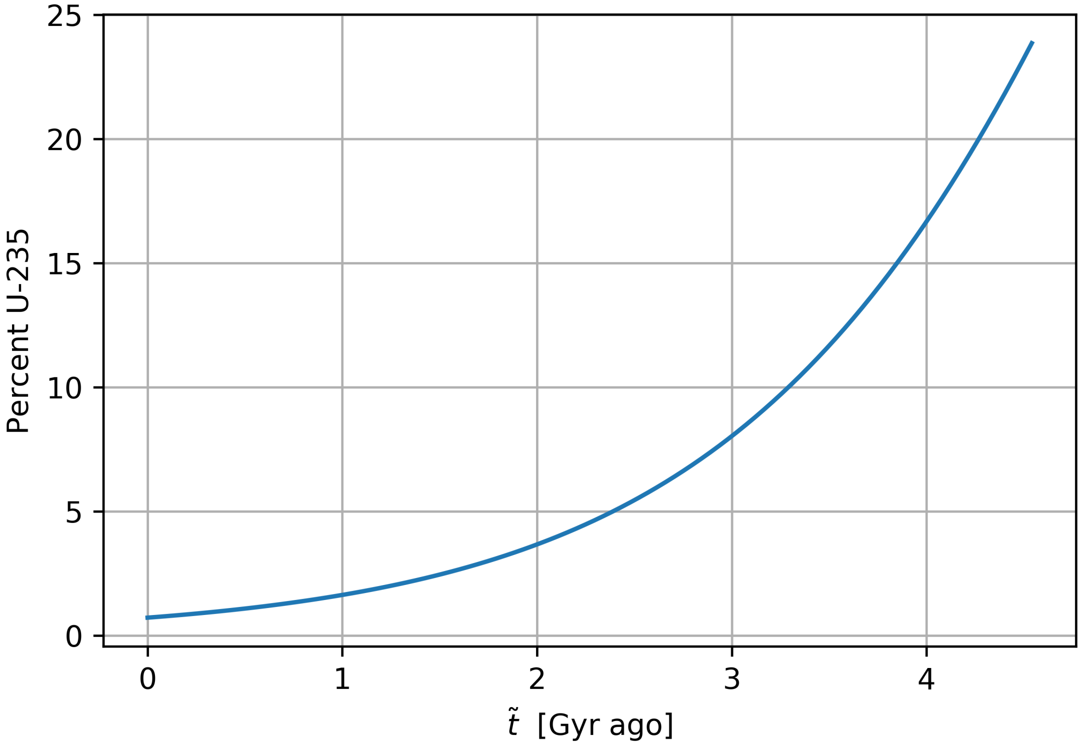

Figure 1: Percent fraction of U-235 in a sample of pure natural Uranium as a function $\tilde{t}$ (Gyr into the past), where $\tilde{t} = 0$ is today with U-235 percent being 0.72%, and $\tilde{t} = 4.5$ is approximately the beginning of the earth.

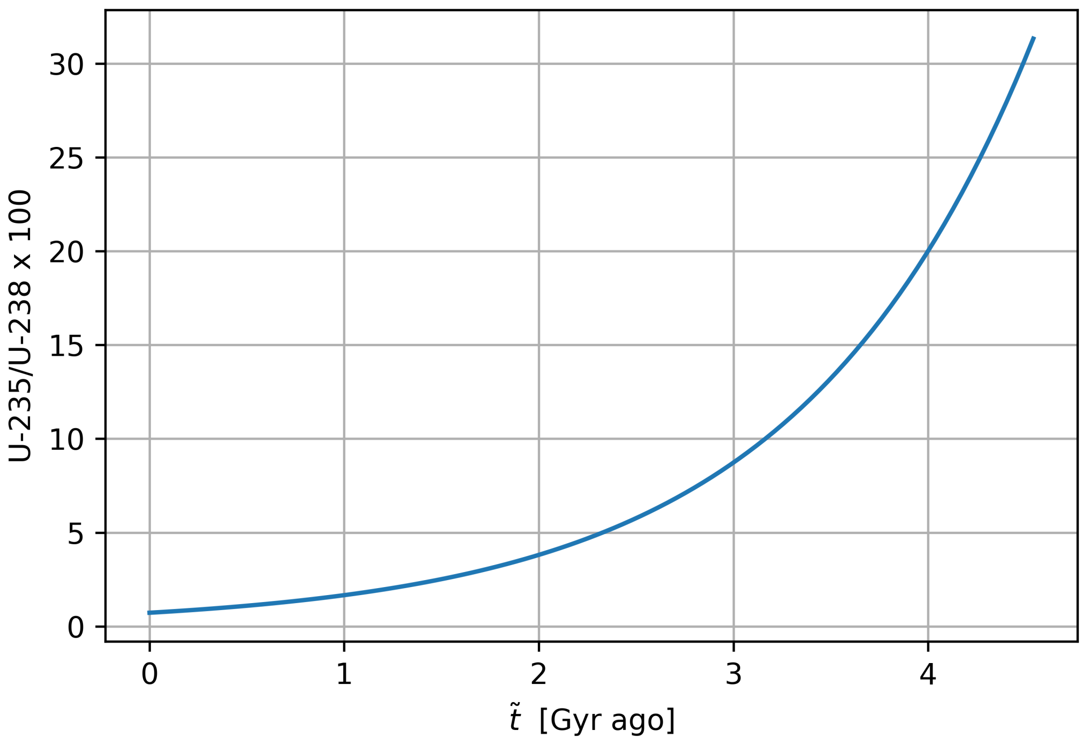

Figure 2: Reproduction of the Gauthier-Lafayete and Weber plot of one hundred times the ratio of U-235 to U-238 in a sample of pure natural Uranium as a function $\tilde{t}$ (Gyr into the past), where $\tilde{t} = 0$ is today and $\tilde{t} = 4.5$ is approximately the beginning of the earth.