ANOMALOUS DEUTERON TO HYDROGEN RATIO IN NATURALLY OCCURRING FISSION REACTIONS AND THE POSSIBILITY OF DEUTERON DISINTEGRATION

M. SHAHEEN, M. RAGHER
University of Illinois at Urbana-Champaign, 163 S. Goodwin Ave., Urbana, IL 61801 USA
(Received June 27, 1971)

A hypothesis is presented for explaining the experimentally determined anomalous D/H ratios observed in the samples from the naturally occurring fission reaction in the Oklo phenomena. No other hypothesis, so far, has been able to explain this data, which is attributed to the unique conditions in the Oklo phenomena. The hypothesis is based on the idea that the Oklo phenomena could be explained by a process of deuteron decay in a closed system of hydrogen, deuterium, helium, and a heavy nucleus. An analysis of the hypothesis is presented, and the results show that the hypothesis is consistent with the experimental data.
ANOMALOUS DEUTERON TO HYDROGEN RATIO IN NATURALLY OCCURRING FISSION REACTIONS AND THE POSSIBILITY OF DEUTERON DISINTEGRATION

M. SHAHEEN, M. RAGHEB

University of Illinois at Urbana-Champaign
185 S. Goodwin Ave., Urbana, IL 61801, USA
(Received June 27, 1961)

A hypothesis is presented for explaining the experimentally determined anomalous D/H ratio observed in the samples from the naturally occurring fissiion reaction in the Oklo phenomenon. No other explanation has been given, to our best knowledge, for the large difference between the measured D/H ratio in the Oklo samples and the expected values in a fission neutron spectrum. A multicomponent system consisting of hydrogen, deuterium, tritium and helium nuclei is considered. An analytical solution is derived and solved using as boundary conditions the experimentally determined value of the D/H ratio. The solution of the rate equations for hydrogen and deuterium concentrations, assuming a pure fission process without a deuteron sink term, yields a D/H ratio of 445 ppm for a reaction in which the flux of neutrons is $10^{15}$ s$^{-1}$. This exceeds the experimentally observed value of 127 ppm, and the naturally occurring value of 150 ppm. Solving the basic rate equations accounting for a deuteron sink term using a hypothesis of deuteron disintegration, and the experimentally observed value of 127 ppm yields a deuteron disintegration constant of $7.47 \times 10^{-21}$ s$^{-1}$. Deuteron disintegration would provide a neutrons source, in addition to the fission neutron, driving a subcritical chain reaction over an extended period of time. Relationship of the presented hypothesis to the Veksler theory of an overabundance anomalous impact explosion explaining the experimentally observed anomalies $235\text{U}/238\text{U}$ ratio, and to the suggestion of deuteron disintegration as a possible explanation of some observations of deuteron disintegration in paladium and titanium electrodes is discussed. The tritium and helium-3 rate equations are further solved under the deuteron disintegration hypothesis and the relationship of the present work to the work by JONES et al. is discussed.

Introduction

The Oklo phenomenon is a process in which a naturally occurring fission chain reaction has taken place. The phenomenon was observed at the Oklo mining site in Gabon, Africa. Samples from the site were extensively studied. Several anomalies in the samples were observed: one pertains to the $235\text{U}/238\text{U}$ ratio, the ratio of the number of fission fragments to the number of fissioned nuclei, another involves the D/H ratio, the atomic ratio of deuterons to hydrogen atoms. The currently accepted explanation for the first of these anomalies is that about 1740 million years ago, when the $235\text{U}$ enrichment was about 3%, instead of the present 0.72%, the conditions at this particular deposit were favorable for a self sustained chain reaction to occur.

* Author to whom correspondence should be addressed.
appropriate conditions were presumably that the geological characteristics at the site allowed the formation of uranium salts soluble in water. Since fission products were found in the samples, it was logical to assume that a fission process had taken place in the ore. Based on this assumption, it is possible to explain the depletion of $^{235}$U and the concentrations of the fission products if the reaction was assumed to have lasted for a period between 600,000 to 1,200,000 years. The fission of neutrons (n/cm$^2$) required to account for these observations, by many different investigators, was estimated to be about $10^{21}$ n/cm$^2$. However, no accepted explanation has been offered for the anomalous D/H ratio.

Assumptions

When samples from the Oklo site were analyzed for water and hydrogen content, it was necessary to heat the samples to about 1160 °C to extract the water from the mineral. This suggests that the water was incorporated into the crystal structure of the mineral. Some uranium ores, in fact, incorporate water, e.g.,

- Carnotite: $K_2(\text{UO}_2)_2(\text{VO}_4)_2 \cdot 3\text{H}_2\text{O}$
- Uranophane: $\text{CaU}_2\text{Si}_2\text{O}_8 \cdot 7\text{H}_2\text{O}$
- Autunite: $\text{Cu}(\text{UO}_2)\text{(PO}_4\text{)}_2 \cdot 10-12\text{H}_2\text{O}$

When the water and hydrogen were analyzed for the D/H ratio, both showed a ratio of about 127 ppm. One particular sample had a D/H ratio, of 127.3 ppm. The D/H ratio in nature is 150 ppm. Furthermore, we will show that in the neutron fluence estimated to explain the uranium-235 depletion in the Oklo samples, the D/H ratio should increase because of neutron capture in hydrogen rather than decrease as implied by the above observations. For a fluence of $10^{22}$ n/cm$^2$, we show that this ratio should be around 445 ppm.

When the water and hydrogen were analyzed for the D/H ratio, both showed a ratio of about 127 ppm. One particular sample had a D/H ratio, of 127.3 ppm. The D/H ratio in nature is 150 ppm. Furthermore, we will show that in the neutron fluence estimated to explain the uranium-235 depletion in the Oklo samples, the D/H ratio should increase because of neutron capture in hydrogen rather than decrease as implied by the above observations. For a fluence of $10^{22}$ n/cm$^2$, we show that this ratio should be around 445 ppm.

Figure 1a shows a multicomponent system describing the major isotopic transitions for elements of interest here in a neutron flux. We consider hydrogen deuterium, tritium and $^3$He concentrations. If we assume that the $^4$He produced will escape from the site and will not contribute further to the isotopic changes, the chain can be simplified to the one shown in Fig 1b. In this multicomponent system, hydrogen can be produced from two sources: (1) $^3$He(n, p) reaction, and (2) postulated deuterium disintegration according to $^2$D $\rightarrow$ n + $^3$H. Deuterons will be produced from neutron capture in hydrogen, or capture of a neutron with production of tritium. Tritium will decay by $\beta^-$ emission to $^3$He with a half-life of 12.3 years. If $^3$He does not escape from the reaction site, it will either undergo an (n, p) or an (n, $\gamma$) reaction. Table 1 shows the cross sections for the different reactions in this chain.

Because of the presence of water, it is justified to assume here that the neutron spectrum is basically thermal. We will further assume that the flux has a Maxwellian shape at room temperature $T_0 = 293$ K. The low reaction rate and the short disintegration time of the $^3$He also justify this assumption. We can then deduce an average cross-section over the Maxwellian spectrum given by $\sigma_\text{w} = 0.886$ assuming that all absorbers are $^3$He.

We shall calculate the initial water concentrations from the data of the sample designated as sample 310, which had a $^{235}$U content of 0.592%. The uranium to ore ratio of this sample was 14.94 weight % and the water content was 9.3%. Based on these data, the initial water concentration $w_0$ can be calculated to be $3.11 \cdot 10^4$ water.
molecule/cm³. Assuming that the water content did not appreciably change during the reaction, we calculate the initial hydrogen and deuterium concentrations as 6.22 × 10²² and 9.33 × 10²³, respectively. This assumption is not necessary as we will show later, since the relevant parameter in the calculations is the ratio R₀ = D₀/H₀ = 150 ppm, the naturally occurring D/H ratio.

The rate equations for the multi-component system shown in Fig. 1, considering a pure fission process without a hypothesis of deuterium disintegration, can be written as:

\[
\begin{align*}
\frac{dH}{dt} &= -\beta_H H + \beta_D H \\
\frac{dD}{dt} &= \beta_H H - \beta_D D \\
\frac{dT}{dt} &= \beta_D D - \lambda_T T + \beta_T T \\
\frac{dHe}{dt} &= \lambda_T T - \beta_T T 
\end{align*}
\]

(1) \hspace{1cm} (2) \hspace{1cm} (3) \hspace{1cm} (4)

where H, D, T, and He are the concentrations in atoms/cm³ of hydrogen, deuterium, tritium, and helium-3, respectively.

We also define:

\[
\begin{align*}
\beta_H &= \alpha_{ch} H (s^{-1}) \\
\beta_D &= \alpha_{ch} D (s^{-1}) \\
\beta_T &= \alpha_{ch} T (s^{-1})
\end{align*}
\]

(5) \hspace{1cm} (6) \hspace{1cm} (7)

where \( \Phi \) is the neutron flux in \( n \cdot \text{cm}^{-2} \cdot \text{s}^{-1} \) and \( \alpha_{ch} \) is the average thermal microscopic capture cross section in \( \text{cm}^² \), we made the approximation:

\[
\alpha_{ch} = \alpha_{ch0}
\]

(8)

implying that the absorptions in \( ^3\text{He} \) are mainly due to the \((n, p)\) reaction which has a large cross-section value as shown in Table 1.

It will be shown later that the flux at Oklo was \(-10^7 n \cdot \text{cm}^{-2} \cdot \text{s}^{-1}\) and since the hydrogen concentration is \(-10^{22}\), it is reasonable to neglect the production of hydrogen from radiolysis and from fission product decay (because of the low flux and hence relatively low fission products concentration), as well as from the \( ^3\text{He} \) \((n, p)\) reaction. The latter approximation is justified by the fact that the concentration of \( ^3\text{He} \) is too low to affect the hydrogen concentration and because \( ^3\text{He} \) is likely to escape from the site of the reaction before any further interactions.

As a first step, we are interested in the H and D concentrations, thus we seek analytical solutions for Eqs (1) and (2), rewritten according to the previously discussed approximations as:

\[
\begin{align*}
\frac{dH}{dt} &= -\beta_H H \\
\frac{dD}{dt} &= \beta_H H - \beta_D D
\end{align*}
\]

(9)

The solution for Eq. (9) is:

\[
H = H_0 e^{-\beta_H t} \\
D = D_0 e^{-\beta_D t} + \frac{\beta_H H_0}{\beta_D - \beta_H} - \frac{\beta_H H_0}{\beta_D - \beta_H} e^{-\beta_D t}
\]

(10)

where \( H_0 \) is the initial hydrogen concentration at time \( t = 0 \).

For future analysis we rewrite this equation in the form:

\[
H = H_0 e^{-\beta_H t} + \frac{\beta_H H_0}{\beta_D - \beta_H} - \frac{\beta_H H_0}{\beta_D - \beta_H} e^{-\beta_D t}
\]

(11)

where \( \Phi = \Phi_0 \) is the fluence in \( \text{n/cm}^² \).

The solution for the linear ordinary differential equation 6 is:

\[
D(t) = \left(D_0 + \frac{\beta_H H_0}{\beta_D - \beta_H} e^{-\beta_H t} - \frac{\beta_H H_0}{\beta_D - \beta_H} e^{-\beta_D t}ight)
\]

(12)

We obtain the ratio \( R = D/H \) in the case of no deuterium disintegration by dividing Eq. (8) by Eq. (9):

\[
R = \left[D_0 + \frac{\beta_H H_0}{\beta_D - \beta_H} e^{-\beta_H t} - \frac{\beta_H H_0}{\beta_D - \beta_H} e^{-\beta_D t}ight] = \left[D_0 + \frac{\beta_H H_0}{\beta_D - \beta_H} e^{-\beta_H t} - \frac{\beta_H H_0}{\beta_D - \beta_H} e^{-\beta_D t}ight]
\]

(13)

where \( D_0 = D_0 / H_0 = 150 \text{ ppm} \).

We notice that \( R \) is a function of the fluence only, since all other parameters are constants. Using the value of the fluence \( \Phi = 10^{22} \text{n/cm}^² \) we get \( R = 445 \text{ ppm} \) which is the R ratio that is expected under the natural reactor assumption.

Figure 2 shows the variation of the ratio \( R = D/H \) as a function of the fluence. We notice that \( R \) remains as a constant \( R_0 \) up to a value of the fluence \( \Phi = 10^{22} \), after which it rises sharply with increasing fluence.

The fact that the D/H ratio observed in the Oklo samples (127 ppm) is much lower than \( R_0 \) (150 ppm), when in fact it is expected to have risen to around 445 ppm as calculated above, is in direct contradiction with the expectations based on the natural reactor assumption alone. It is an anomaly that we propose to explain here.

We first assume that the anomaly is due to a nuclear process since we are unaware of any chemical or other physical processes which could have depleted the deuterium and/or enriched the hydrogen content of the ore mineral. To explain this anomaly, we investigate the possibility that the deuterons have undergone some process of
disappearance or disintegration. In this regard, one aspect of the natural self-sustained fission chain reaction hypothesis that needs to be modified is that the core itself formed a critical assembly, and that the reaction was sustained only by the fission neutrons source $S$. Regardless of the mechanism involved, if we adopt the deuteron disintegration hypothesis, the following reaction would take place:

$$^1D^2 \rightarrow ^1H + ^2He$$

The neutrons produced would act as an additional source $S_d$ that will further drive the reaction through the combined source:

$$S = S_t + S_d$$

If the deposit is to remain critical in the presence of the extra neutrons from deuteron disintegration, the core must have been subcritical. We will show later that the multiplication factor of the core assembly could have been around 0.998, a subcritical configuration.

**Rate equations assuming deuteron disintegration**

As we write the rate equations for hydrogen and deuterium, neglecting the production of hydrogen from radiolysis and the $^9Be(n, p)$ reaction, and account for the possible deuteron disintegration through a sink term of the form $\lambda_2D$, where $\lambda_2$ is the deuteron disintegration constant (s$^{-1}$), then we obtain:

$$\frac{dH}{dt} = \lambda_2D - \beta_HH$$  \hspace{1cm} (11)

$$\frac{dD}{dt} = \beta_HH - (\beta_D + \beta_N)D$$  \hspace{1cm} (12)

This is a system of two coupled first order linear ordinary differential equations. An analytical solution is possible by means of the Laplace transformation (L. T.) method. Using the notation:

$$L[H(t)] = h(s)$$

and

$$L[D(t)] = d(s)$$

where $L$ is the Laplace transformation operator.

Using the L. T. property:

$$L[f(t)g(t)] = sL[f(t)] - f(0)$$

and taking the L. T. of both sides of Eqs (11) and (12), we can write the rate equations

for $H$ and $D$ in the s-domain, transforming the differential equation system into a system of algebraic equations:

$$sh(s) - H_s = \lambda_2h(s) - \beta_Hh(s)$$  \hspace{1cm} (13)

$$sd(s) - D_s = \beta_Hh(s) - (\beta_D + \beta_N)d(s)$$  \hspace{1cm} (14)

where $D_s$ and $H_s$ are the initial concentrations of deuterium and hydrogen, respectively.

By solving Eqs (13) and (14) for $d(s)$, we obtain:

$$d(s) = \frac{sD_0 + \beta_HH_0 + D_0}{s + (\beta_H + \beta_D + \lambda_2)s + \beta_N\beta_D}$$  \hspace{1cm} (15)

To get the inverse L. T. of $d(s)$ we use the partial fraction method. To do so, we have to write the denominator as a product of its primary factors. Let:

$$s^2 + (\beta_H + \beta_D + \lambda_2)s + \beta_N\beta_D = (s - s_1)(s - s_2)$$

where:

$$s_1 = \frac{1}{2}(\beta_H + \beta_D + \lambda_2) - \frac{1}{2}\sqrt{(\beta_H + \beta_D + \lambda_2)^2 - 4\beta_N\beta_D}$$

$$s_2 = \frac{1}{2}(\beta_H + \beta_D + \lambda_2) + \frac{1}{2}\sqrt{(\beta_H + \beta_D + \lambda_2)^2 - 4\beta_N\beta_D}$$

and the choice of whether $s_1$ or $s_2$ should be added to or subtracted from the square root term is arbitrary.

Further, we know that:

$$s_1 + s_2 = -\beta_D$$

$$s_1s_2 = \beta_N\beta_D$$

Letting $N_0 = H_0 + D_0$, we now write $d(s)$ as:

$$d(s) = \frac{sD_0 + \beta_HN_0}{(s - s_1)(s - s_2)}$$  \hspace{1cm} (16)

Separating into partial fractions, we get:

$$d(s) = \frac{sD_0 + \beta_HN_0}{(s - s_1)(s - s_2)} + \frac{sD_0 + \beta_HN_0}{(s - s_2)(s - s_1)}$$  \hspace{1cm} (17)
From the properties of the inverse L. T., we can write for the inverse transformation:

\[ L^{-1} \left[ b \right] = \frac{b}{s-a} \cdot e^{as} \]

Taking the inverse L. T. of Eq. (20), we have:

\[ D(s) = \frac{s_1 D_0 + \beta_0 N_0}{s_1 - s_2} \cdot e^{s_1 - s_2} + \frac{s_2 D_0 + \beta_0 N_0}{s_2 - s_1} \cdot e^{s_2 - s_1} \]

where \( s_1 \) and \( s_2 \) are defined by Eq. (16).

To obtain \( H(t) \), from Eq. (13):

\[ h(s) = \frac{H_0}{s + \beta_0} + \frac{\lambda_D}{s} \cdot \frac{d(s)}{s + \beta_0} \]

Substituting for \( d(s) \) from Eq. (20), we get:

\[ h(s) = \frac{H_0}{s + \beta_0} + \frac{\lambda_D}{s} \left[ \frac{s_1 D_0 + \beta_0 N_0}{s_1 - s_2} \cdot e^{s_1 - s_2} + \frac{s_2 D_0 + \beta_0 N_0}{s_2 - s_1} \cdot e^{s_2 - s_1} \right] \]

Using the inverse L. T. tables:

\[ L^{-1} \left[ \frac{1}{(s-a)(s-b)} \right] = \frac{1}{a-b} \left( e^{as} - e^{bs} \right) \]

Taking the inverse L. T. of Eq. (23) and rearranging, we get:

\[ H(t) = \lambda_D \left[ \frac{s_1 D_0 + \beta_0 N_0}{s_1 - s_2} \cdot e^{s_1 - s_2} + \frac{s_2 D_0 + \beta_0 N_0}{s_2 - s_1} \cdot e^{s_2 - s_1} \right] + \lambda_D \left[ \frac{H_0}{s + \beta_0} \cdot e^{-\beta_0 t} \right] \]

It is possible to use Eqs (17) and (18) to prove that the term multiplying \( e^{-\beta_0 t} \) is equal to zero. Therefore, Eq. (25) simplifies to:

\[ H(t) = \lambda_D \left[ \frac{s_1 D_0 + \beta_0 N_0}{s_1 - s_2} \cdot e^{s_1 - s_2} + \frac{s_2 D_0 + \beta_0 N_0}{s_2 - s_1} \cdot e^{s_2 - s_1} \right] \]

We should note here that Eqs (21) and (26) tend to the limit to Eqs (9) and (7), which were derived in the case of no deuteron disintegration, in the special case of \( \lambda_D = 0 \). From Eq. (16) we find that when \( \lambda_D = 0 \), then \( s_1 = -\beta_0 \) and \( s_2 = -\beta_D \). It is straightforward to prove that Eq. (21) reduces to Eq. (9) when \( \lambda_D = 0 \). However, this is not the case for Eq. (26). It might seem at first glance that when \( \lambda_D = 0 \) the equation reduces to \( H(t) \rightarrow 0 \) in contradiction to Eq. (7). However, careful consideration will reveal that the solution tends to the undefined value 0.0. We shall evaluate it using L'Hopital's Rule. Taking the limit of \( H(t) \) as \( \lambda_D \rightarrow 0 \) and hence \( s_1 \rightarrow -\beta_0 \), we get:

\[ \lim_{\lambda_D \rightarrow 0} H(t) = \lim_{s_1 \rightarrow -\beta_0} \left[ \frac{\lambda_D s_1 D_0 + \beta_0 N_0}{s_1 + \beta_0} \cdot e^{-\beta_0 t} + \frac{\lambda_D}{s_1 + \beta_0} \cdot e^{-s_1 t} \right] \]

The second term tends to zero without problems. The first term tends to 0.0. Using Eq. (16a), we express \( s_1 \) in terms of \( \lambda_D \) as:

\[ \lim_{\lambda_D \rightarrow 0} \left[ \frac{\lambda_D s_1 D_0 + \beta_0 N_0}{s_1 + \beta_0} \cdot e^{-\beta_0 t} \right] = \frac{-\lambda_D \beta_0 H_0 e^{-\beta_0 t}}{(s_1 + \beta_0)(1/2)} \]

Differentiating the denominator and the numerator with respect to \( \lambda_D \) and then setting \( \lambda_D = 0 \), we get:

\[ H(t) \lim_{\lambda_D \rightarrow 0} \left[ \frac{-\lambda_D \beta_0 H_0 e^{-\beta_0 t}}{(s_1 + \beta_0)(1/2)} \right] = H_0 e^{-\beta_0 t} \]

which is the same result as earlier obtained in Eq. (7).

Expression for the deuteron to hydrogen D/H ratio

Dividing Eq. (21) by Eq. (26) to obtain \( D(t)/H(t) \), which we denote as \( R(t) \), we obtain:

\[ R(t) = \frac{1}{\lambda_D} \left[ \frac{(s_1 D_0 + \beta_0 N_0)e^{s_1 t} - (s_2 D_0 + \beta_0 N_0)e^{s_2 t}}{(s_1 + \beta_0)(1/2)} \right] \]
Dividing both the numerator and the denominator by $N_0$ and noting that $N_0 = H_0 + D_0$, we have:

$$D_0/N_0 = D_0/H_0 = R_0/(1 + R_0),$$

where $R_0 = D_0/H_0 = 150$ ppm.

Then $1 + R_0 = 1 + 1.5 \cdot 10^{-4} = 1$.

Consequently, $D_0/N_0 = R_0$, and Eq. (27) reduces to:

$$R(t) = \frac{1}{\lambda_0} \left[ \frac{(s_2R_0 + \beta_0)t^{s_2} - (s_2R_0 + \beta_0)t^{s_2-1} - (s_2R_0 + \beta_0)t^{s_2-2} - \ldots}{(s_2 + \beta_0)} \right]$$

(28)

It is observed here that the initial water content terms, i.e., $H_0$ and $D_0$, do not appear in the equation but rather the ratio $R_0 = D_0/H_0$. This means that the results based on the Oklo Sample #310, in which the water content was 9.3%, do not really depend on the sample and that we could have obtained the same results for any other sample that had undergone the Oklo phenomenon. It is interesting to note here that the D/H ratios observed in the Oklo phenomenon (which we denote as $R_0$) for Sample KBN47 and Sample #556 were 131.2 and 126.1, respectively, even though those samples had water contents of 13.2% and 8.5%, respectively.

For an Oklo reaction that has lasted for time $t_0$, the final D/H ratio, $R_0$, is then given by:

$$R_0 = \frac{1}{\lambda_0} \left[ \frac{(s_2R_0 + \beta_0)t^{s_2} - (s_2R_0 + \beta_0)t^{s_2-1} - (s_2R_0 + \beta_0)t^{s_2-2} - \ldots}{(s_2 + \beta_0)} \right]$$

(29)

where both $s_1$ and $s_2$ are functions of $\lambda_0$.

It turns out that Eq. (29) is nonlinear, and a solution can be sought in the form:

$$\lambda_0^{s_1} = F(R_0, \Phi, t_0, \lambda_0)$$

which implies that to obtain $\lambda_0$ for a given $R_0$, $\Phi$, and $t_0$, we need to use an iterative approach. To obtain the complete functional dependence in Eq. (29), let us identify the independent and dependent parameters. There are three independent parameters: the fluence, $\Phi$, the time duration $t_0$, and the isogenic disintegration constant $\lambda_0$. The flux, $\Phi = \phi(t)$, and $\beta_0$, $\beta_0$ (the product of the cross section and the flux), and $s_1$, $s_2$ are functions of $\beta_0$, $t_0$, and $\lambda_0$, i.e., $s$ and $\lambda_0$.

Independent parameters:
- $\Phi$, $t_0$, $\lambda_0$

Results

We calculated the value of the deuteron disintegration constant $\lambda_0$ under the conditions of the Oklo phenomenon for three values of $\Phi$ around $10^{21}$: $0.5 \cdot 10^{21}$, $1.0 \cdot 10^{21}$, and $1.5 \cdot 10^{21}$, and for different time durations in the range: $3 \cdot 10^6$–$1.2 \cdot 10^9$ years. Although the variation of the estimated time duration is large, the value of $\lambda_0$ calculated for the same fluence and for the same limiting time periods is not very large. Similarly, for the same time duration the variation of $\lambda_0$ for fluence values $0.5 \cdot 10^{21}$ and $1.5 \cdot 10^{21}$ is also not large. We adopt as a reference case that of $\Phi = 10^{21}$ and $t_0 = 10^6$ years.
First we show in Fig. 3 the variation of the flux with different time durations over the three fluence cases. For the reference case, the flux would be $3.17 \cdot 10^7 \text{n cm}^{-2} \text{s}^{-1}$ which meets the condition that:

$$\phi \ll \lambda_{239Pu}/(\alpha_{239Pu})$$

stated in Reference 1. In Fig. 4 we show the D/H ratio, $R$, for the range of $\lambda_0$ values

between $10^{-29}$ and $10^{-6} \text{ s}^{-1}$ for the three fluence cases for $t_d = 10^6$ years. The value of $R$ at $\lambda_0 = 10^{-29}$ is zero which is equal to that from Fig. 2. For the same fluence, $R$ drops to zero at values greater than $10^{-9}$ and the variation is negligible at $\lambda_0$ less than $10^{-13}$.

One can compute $\lambda_{239Pu}$, i.e., the value of $\lambda_0$ that would lead to a D/H ratio $R = R_b = 127.3$ from the sample Oklo #31U for a reaction in which the fluence was $\Phi$ and have lasted for time $t_d$. For the reference case, this turns out to be $7.47 \cdot 10^{-20} \text{ s}^{-1}$.

Figure 5 shows the variation of $R$ with $\lambda_0$ for $\Phi = 10^{22}$ for different time durations. At $t_d = 6 \cdot 10^6$ $\text{yr}$, $\lambda_0 = 1.24 \cdot 10^{-13}$ and for $t_d = 10^6$, $\lambda_0 = 7.47 \cdot 10^{-14} \text{ s}^{-1}$. Figure 6 shows the variation of $\lambda_{239Pu}$ with the time period of the Oklo reaction for the three fluence cases.

If we assume that deuteron disintegration is not a single particle reaction but rather a multiple particle interaction, i.e., that the deuteron disintegration is not similar to the natural decay process, but rather the deuteron disintegrates whenever it exists under certain conditions created by the system that consists of the deuteron itself and the surrounding atoms, whether these atoms are other deuterons or heavier atoms, then the deuteron disintegration reaction may proceed in one of two ways. As a free neutron reaction:

$$ D^2 + 2N^\prime \rightarrow H^1 + p^1 + 2N^\prime $$

(30)
here the nucleus $^2N^A$ acts as a catalyst for the deuteron disintegration process, and does not undergo any isotopic changes. However, another more likely way for the reaction to proceed which leads to isotopic changes in the medium in which the deuteron disintegration reaction occurs is the one in which the deuteron gives up its neutron to the nucleus $^2N^A$ according to:

$$D^2 + ^2N^A \rightarrow ^{1}$H + $^2N^{A-1}$ \tag{31}$$

An example of this kind of reaction would be:

$$D^2 + ^2D^2 \rightarrow ^3P + ^2H^1$$

$$^3P \rightarrow ^2P^0 + ^2He^3$$

Provided that the reaction time is long enough to allow for complete decay of the tritium to helium-3, we can write the net reaction as:

$$D^2 + ^2D^2 \rightarrow ^2P^0 + ^2H^1 + ^2He^3 \tag{32}$$

Compare this to the same reaction proceeding by neutron capture:

$$D^2 + ^1n \rightarrow ^2P$$

$$^2P \rightarrow ^2P^0 + ^2He^3$$

Again, if the reaction proceeds for a long enough time, then the net reaction is:

$$D^2 + ^1n \rightarrow ^2P^0 + ^2He^3 \tag{33}$$

Both reactions involve the depletion of deuterons with the production of tritium and $^3He$. However, the first reaction involves the depletion of twice as many deuterons and also leads to the production of protons. If an experiment is performed in which the Oklo conditions are reproduced, the hypothesis can be tested based on the outcome of reactions 32 or 33.

According to this assumption, we can state that the rate of deuteron disintegrations is proportional to the product of deuteron concentration $D$ and the concentration of the host atom $N$, i.e.:

$$\lambda_D D \propto D \cdot N \tag{34}$$

We further assume that the concentration of the host atom, which would be uranium in the Oklo reaction, is constant. This assumption is valid to a great extent since the uranium depletion in the estimated fluence will be $10^{-3}$ of the initial uranium concentration. Denoting the proportionality constant in Eq. (34) by $\mu_D$, we obtain:

$$\lambda_D = \mu_D N$$

$$\mu_D = \lambda_D N \text{ cm}^{-3} \text{s}^{-1} \tag{35}$$

We note that $\mu_D$ has the same units as $<\sigma V>$, the reaction parameter used in plasma physics and fusion studies.

If the reaction was a D-U disintegration reaction, i.e., deuterons disintegrating while present in the uranium lattice, then from the sample Oklo #310 data, and assuming that the mineral was in the form of $UO_2$, the U concentration would be $3.33 \cdot 10^{21}$ U atoms/cm$^3$ and:

$$\mu_{DU} = \lambda_D/U = 2.24 \cdot 10^{-35} \text{ cm}^{3} \text{s}^{-1}$$

However, the result is not valid for a D-D disintegration reaction as the term describing the deuteron disintegration in this case should be $\lambda D^2$. We have accounted for this situation in the numerical calculation scheme and the D-D disintegration constant that will account for the $D/2H$ ratio is found to be $-10^{-31}$ cm$^3$/s. If the reaction is a D-D disintegration reaction that can be described by the term $\lambda_D D$, then the reaction constant would be $7.47 \cdot 10^{-14}$, [deuteron disintegrations/deuteron - s].
M. SHAHEEN, M. RAGHEB: ANOMALOUS DEUTERON TO HYDROGEN RATIO

Tritium and helium-3 concentrations

As shown in Fig. 1, the Oklo reaction will lead to the production of T and He. To obtain their concentrations, the rate equations (3) and (4) should be solved with D(t) given by Eq. (21). The rate equations are:

\[ \frac{dT}{dt} = \beta_D D(t) - \lambda_T T + \beta_p \text{He} \]  
(36)

\[ \frac{d\text{He}}{dt} = \lambda_T T - \beta_p \text{He} \]  
(37)

We shall solve first for the case where \(^3\text{He}\) does not escape from the site of the reaction and shall account for the \(^3\text{He} (n, p)\) reaction. We shall obtain the case where \(^3\text{He}\) leaks out of the site as a special case when \(\beta_p = 0\). We shall show that the concentration of \(^3\text{He}\) remains unchanged in both cases. Equations (36) and (37) are two coupled linear ordinary differential equations which will be solved by the L. T. method in exactly the same manner used to solve for the H and the D concentrations. Hence the details of solution will not be shown. For the case where the initial concentrations of T and \(^3\text{He}\) are zero and \(^3\text{He}\) does not leak out, we get:

\[ T(t) = N_0 \beta_D \left\{ \frac{(s_1 + \beta_D)(s_2 + \beta_D)}{s_1(s_2 - s_3)} e^{s_1 t} + \frac{(s_1 + \beta_D)(s_2 + \beta_D)}{s_2(s_3 - s_2)} e^{s_2 t} \right\} + \frac{(s_1 + \beta_D)(s_2 + \beta_D)}{s_3(s_3 - s_2)(s_3 - s_3)} e^{s_3 t} - \frac{\beta_p}{\beta_p} \]  
(38)

and

\[ \text{He}(t) = N_0 \lambda_T \beta_D \left\{ \frac{(s_1 + \beta_D) s_2}{s_1(s_2 - s_3)(s_2 - s_3)} e^{s_1 t} - \frac{(s_1 + \beta_D) s_2}{s_2(s_3 - s_2)(s_3 - s_3)} e^{s_2 t} - \frac{1}{\beta_p s_3} \right\} \]  
(39)

where \(\lambda_T\) is the tritium disintegration constant and \(s_3 = -(\beta_p + \lambda_T)\).

It is possible to obtain the solution in the case where the initial concentrations of T and \(^3\text{He}\) are not zero. However, there is no reason to assume that T or \(^3\text{He}\) existed in the ore initially, and hence we will not show the solution in that case. We note from Eqs (38) and (39) that the concentrations of T and \(^3\text{He}\) are proportional to the initial water content in the rock. For the typical case of \(\Phi = 10^5, \quad \chi = 10^4\) so that \(\lambda_T = 7.47 \cdot 10^{-14}\) the change in T and He concentrations with time is shown in Fig. 7. The final concentrations in this case are \(T(t) = 3.6 \cdot 10^7\) and \(\text{He}(t) = 3.78 \cdot 10^{10}\). The rate of \(^3\text{He}\) production would be \(\text{He}(t) \lambda_T = 0.012 \text{ He atom/cm}^3\cdot\text{s}\).

Fig. 7. Variation of tritium and helium-3 concentrations with time

If \(^3\text{He}\) escapes from the site of the reaction, then it will not further interact to produce H and T in the \((n, p)\) reaction and the rate equations become:

\[ \frac{dT}{dt} = \beta_D D - \lambda_T T \]  
(40)

\[ \frac{d\text{He}}{dt} = \lambda_T T \]  
(41)

The solution will be given by Eqs (38) and (39) with \(\beta_p = 0\). For the reference case, the final tritium concentration is \(6.34 \cdot 10^6\) at/cm\(^3\) and the total helium-3 production is \(3.78 \cdot 10^{10}\) at/cm\(^3\). If all the helium-3 produced escapes, the rate of \(^3\text{He}\) release to the atmosphere would be \(0.012\) at/cm\(^3\)\cdot\text{s}\).

To explain why the \(^3\text{He}\) concentration does not change when setting \(\beta_p = 0\), we note that \(\beta_p\) appears in Eq. (33) for He within the term \(s_3 = -(\beta_p + \lambda_T)\). For the Oklo cases, \(\beta_p \sim 10^{-15}\) and \(\lambda_T = 1.79 \cdot 10^{-9}\), so that \(\lambda_T >> \beta_p\) and \(s_3 = -\lambda_T\). Hence \(\beta_p\) will not affect the \(^3\text{He}\) concentration.
Explain the anomalous $^{238}\text{U}/^{235}\text{U}$ ratio. According to the proposed model, $^{238}\text{U}$ can undergo the following reactions:

\[
\begin{align*}
1\text{D} + 2^{238}\text{U} & \rightarrow \text{F}_1 + \text{F}_2 + \text{H} + \nu + \pi^1 \\
1\text{D} + 2^{235}\text{U} & \rightarrow \text{H} + 2^{234}\text{U}
\end{align*}
\]

where $\text{F}_1$ and $\text{F}_2$ are fission products and $^{236}\text{U}$ eventually produces $^{239}\text{Pu}$. The same two reactions are possible with $^{235}\text{U}$. Deuteron disintegration would produce a source of neutrons $S$, the number of such neutrons will be given by $S_{\text{D}}$. For the typical case, if we take the average value of the deuteron concentration to be $(\text{D}_1 + \text{D}_2)/2$, then the number of neutrons will be equal to $6.44 \cdot 10^9$ fast neutrons/(cm$^2$·s). If we denote the multiplication factor of the assembly by $k$, then the flux is given by:

\[
\phi = S/(1 - k).
\]

For $k$ less than unity, or for a subcritical system:

\[
\phi = S/(1 - k).
\]

Since $\phi = 3.17 \cdot 10^7$, and $S = 6.44 \cdot 10^9$, thus $k = 0.999$, implying a subcritical, source-driven system.

JONES et al.\(^5\) suggest that cold nuclear fusion takes place in the earth interior. Based on $^{3}\text{He}$ flow out of the earth mantle of $2 \cdot 10^{19}$ $^{3}\text{He}$ atoms/s, and that the mantle water reservoir is $1.4 \cdot 10^{24}$ g, they estimate a value for the $\text{D} + \text{D}$ fusion constant of $10^{29}$ fusion/deuteron·s. The value which we calculated for a $\text{D} + \text{D}$ disintegration reaction in a high $Z$ medium is $7.47 \cdot 10^{14}$ deuteron disintegration/deuteron·s, which is about ten orders of magnitude higher than that calculated by JONES. It is interesting to note that this ratio is close to the ratio of the OPPENHEIMER-PHILLIPS to the GAMOW penetrabilities of the potential barrier of a high $Z$ nucleus to a low $Z$ nucleus by low energy deuterons, which is about $10^8$.\(^6\)

We can estimate, according to our hypothesis, the $^{3}\text{He}$ flow out of the earth's mantle. We have calculated that under the localized conditions of the Oklo deposit, the rate of $^{3}\text{He}$ escape out of the deposit is $0.012$ $^{3}\text{He}$ atom/cm$^2$·s. If we assume that the same conditions that existed at Oklo prevail in the mantle of the earth, and if we assume that the mantle water density is $1$ g/cm$^3$ so that the mantle volume is $1.4 \cdot 10^{27}$ cm$^3$ then we can calculate the total $^{3}\text{He}$ flow out of the earth mantle due to a reaction similar to the Oklo reaction to be $1.7 \cdot 10^{21}$ $^{3}\text{He}$ atoms. This can be compared to JONES's value of $2 \cdot 10^{19}$ keeping in mind that the conditions of the Oklo reaction may have been more favorable to the deuteron disintegration reaction than currently exist in the earth's mantle as discussed by JONES.\(^5\)

Deuteron disintegration may be considered as a possible explanation of some
observation of deuterium reactions in palladium and titanium electrodes. The phenomenon is observed in lattices of these elements loaded with deuterium, the host lattice will be element $N$ in Eqs (30) or (31). Neutron production from deuteron disintegration would lead to isotopic changes. The protons released are not likely to be absorbed and may be detected. Tritium production is possible from a D-D deuteron disintegration reaction in which a neutron from the disintegration of one deuteron is absorbed in another deuteron. Further investigation of the Oklo data may help generate a better understanding of these observations.

References