SEPARATUM

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

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(Received June 27, 1991)

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Introduction

The Oklo phenomenon is a process in which a naturally occurring fission chain reaction has taken place.[1] 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 $^{235}U/^{238}U$ 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}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.[1] The

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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.[2] The fluence of neutrons (n/cm^2) required to account for these observations, by many different investigators,[1,3,4] 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.[4] This suggests that the water was incorporated into the crystal structure of the mineral. Some uranium ores, in fact, incorporate water, e.g.,

Carnotite: $K_2(UO_2)_2(VO_4)_2 \cdot 3H_2O$

Uranophane: $CaU_2Si_2O_{11} \cdot 7H_2O$

Autunite: $Ca(UO_2)_2(PO_4)_2 \cdot 10\text{–}12H_2O$

When the water and hydrogen were analyzed for the D/H ratio, both showed a ratio of about 127 ppm.[4] 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^{21} 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 ^3He concentrations. If we assume that the ^3He 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) $^3He(n, p)$ reaction, and (2) postulated deuteron disintegration according to: ${}^2_1D \rightarrow {}^1_0n + {}^1_1H$. Deuterons will be produced from neutron capture in hydrogen,[5,6] or capture of a neutron with production of tritium. Tritium will decay by β^- emission to ^3He with a half-life of 12.26 years. If ^3He does not escape

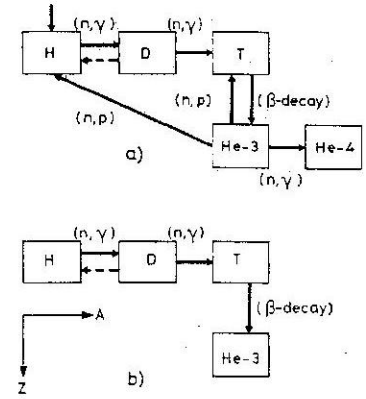

Fig. 1. Multicomponent system consisting of hydrogen, deuterium, tritium, and helium-3, and isotopic transitions of elements in a neutron flux

Table 1
Thermal cross sections for H, D, T, and ^3He neutron reactions[16]

Reaction	Thermal neutron cross section, barn
$H(n, \gamma) D$	0.333
$D(n, \gamma) T$	$0.51 \cdot 10^{-3}$
$T(n, \gamma) H^4$	$< 6 \cdot 10^{-3}$
$3He(n, p) T$	5330
$^3He(n, \gamma) He^4$	0.05

from the reaction site, it will either undergo an (n, p) or an (n, γ) 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 heat dissipation from the ore also justify this assumption. We can then deduce an average cross-section over the Maxwellian spectrum given by $\sigma_{av} = 0.886\,\sigma$ assuming that all absorbers are 1/v absorbers.

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%.[4] Based on these data, the initial water concentration w_0 can be calculated to be $3.11 \cdot 10^{21}$ water

molecule/cm^3. Assuming that the water content did not appreciably change during the reaction, we calculate the initial hydrogen and deuteron concentrations as $6.22 \cdot 10^{21}$ and $9.33 \cdot 10^{17}$, respecctively. This assumption is not necessary as we will show later, since the relevant parameter in the calculations is the ratio $R_0 = D_0/H_0 = 150$ 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 deuteron disintegration, can be written as:

$$dH/dt = -\beta_H H + \beta_p He \quad (1)$$

$$dD/dt = \beta_H H - \beta_D D \quad (2)$$

$$dT/dt = \beta_D D - \lambda_T T + \beta_p He \quad (3)$$

$$dHe/dt = \lambda_t T - \beta_p He \quad (4)$$

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

We also define:

$$\beta_H = \sigma_{\gamma H}\phi, \ (s^{-1})$$

$$\beta_D = \sigma_{\gamma D}\phi, \ (s^{-1})$$

$$\beta_p = \sigma_{pHe}\phi \ (s^{-1})$$

where Φ is the neutron flux in $n \cdot cm^{-2} \cdot s^{-1}$ and σ_γ is the average thermal microscopic capture cross section in cm^2, we made the approximation:

$$\sigma_{pHe} = \sigma_{aHe}$$

implying that the absorptions in ^{3}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 $\sim 10^7$ $n \cdot cm^{-2} \cdot s^{-1}$ and since the hydrogen concentration is $\sim 10^{21}$, 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}He (n, p) reaction. The latter approximation is justified by the fact that the concentration of ^{3}He is too low to affect the hydrogen concentration and because ^{3}He is likely to escape from the site of the reaction before any further interactions.[4]

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:

$$dH/dt = -\beta_H H \quad (5)$$

$$dD/dt = \beta_H H - \beta_D D \quad (6)$$

The solution for Eq. (5) is:

$$H = H_0 e^{-\beta_H t} \quad (7)$$

where H_0 is the initial hydrogen concentration at time $t = 0$.

For future analysis we rewrite this equation in the form:

$$H = H_0 e^{-\sigma_{\gamma H}\phi t} = H_0 e^{-\sigma_{\gamma H}\Phi} \quad (8)$$

where $\Phi = \phi t$ is the fluence in n/cm^2.

The solution for the linear ordinary differential equation 6 is:

$$D(t) = \left\{D_0 + \frac{\beta_H H_0}{\beta_H - \beta_D}\right\} e^{-\beta t/D} - \left\{\frac{\beta_H H_0}{\beta_H - \beta_D}\right\} e^{-\beta_H t} \quad (9)$$

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

$$R = \left\{R_0 + \frac{\sigma_{\gamma H}}{\sigma_{\gamma H} - \sigma_{\gamma D}}\right\} e^{(\sigma_{\gamma H} - \sigma_{\gamma D})\Phi_i} - \left\{\frac{\sigma_{\gamma H}}{\sigma_{\gamma H} - \sigma_{\gamma D}}\right\}$$

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

We notice that R is a function of the fluence only, since all other parameters are constants. Using the value of the fluence $\Phi = 10^{21}$ n/cm^2, we get $R = 445$ 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 $\Phi = 10^{20}$, after which it rises sharply with increasing fluence.

The fact that the D/H ratio observed in the Oklo samples (127 ppm) is reduced below 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 ore itself formed a critical assembly, and that the reaction was sustained only by the fission neutrons source: S_f. Regardless of the mechanism involved, if we adopt the deuteron disintegration hypothesis, the following reaction would take place:

$$ {}_1D^2 \rightarrow {}_1H^1 + {}_0n^1. $$

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

$$ S = S_f + S_d. $$

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

Rate equations assuming deuteron disintegration

It we write the rate equations for hydrogen and deuterium, neglecting the production of hydrogen from radiolysis and the ^3He (n, p) reaction, and account for the possible deuteron disintegration through a sink term of the form $\lambda_D D$, where λ_d is the deuteron disintegration constant (s^{-1}), then we obtain:

$$ dH/dt = \lambda_D D - \beta_H H \tag{11} $$

$$ dD/dt = \beta_H H - (\lambda_D + \beta_D) D \tag{12} $$

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:

$$ L\,dF(t)/dt = s\,L\,F(t) - F(0), $$

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:

$$ sh(s) - H_0 = \lambda_D d(s) - \beta_H h(s) \tag{13} $$

$$ sd(s) - D_0 = \beta_H h(s) - (\lambda_D + \beta_D) d(s) \tag{14} $$

where D_0 and H_0 are the initial concentrations of deuterium and hydrogen, respectively.

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

$$ d(s) = \frac{sD_0 + \beta_H(H_0 + D_0)}{s^2 + (\beta_H + \beta_d + \lambda_D)s + \beta_H\beta_D} \tag{15} $$

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:

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

where:

$$ s_1 = 1/2\{ -(\beta_H + \beta_D + \lambda_D) - [(\beta_H + \beta_D + \lambda_D)^2 - 4\beta_H\beta_D]^{1/2}\} \tag{16a} $$

$$ s_2 = 1/2\{ -(\beta_H + \beta_D + \lambda_D) + [(\beta_H + \beta_D + \lambda_D)^2 - 4\beta_H\beta_D]^{1/2}\} \tag{16b} $$

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:

$$ s_1 + s_2 = -(\beta_H + \beta_D + \lambda_D) \tag{17} $$

$$ s_1 \cdot s_2 = \beta_H\beta_D \tag{18} $$

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

$$ d(s) = \frac{sD_0 + \beta_H N_0}{(s - s_1)(s - s_2)} \tag{19} $$

Separating into partial fractions, we get:

$$ d(s) = \frac{s_1 D_0 + \beta_H N_0}{(s_1 - s_2)(s - s_1)} + \frac{s_2 D_0 + \beta_H N_0}{(s_2 - s_1)(s - s_2)} \tag{20} $$

From the properties of the inverse L. T., we can write for the inverse transformation:

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

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

$$D(t) = \frac{s_1 D_0 + \beta_H N_0}{(s_1 - s_2)} e^{s_1 t} + \frac{s_2 D_0 + \beta_H N_0}{(s_2 - s_1)} e^{s_2 t} \tag{21}$$

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

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

$$(s + \beta_H)h(s) - \lambda_D d(s) = H_0$$

$$h(s) = \frac{H_0 + \lambda_D d(s)}{(s + \beta_H)} = \frac{H_0}{(s + \beta_H)} + \lambda_D \frac{d(s)}{(s + \beta_H)} \tag{22}$$

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

$$h(s) = \frac{H_0}{(s + \beta_H)} + \lambda_D \left\{ \frac{s_1 D_0 + \beta_H N_0}{(s_1 - s_2)(s - s_1)(s + \beta_H)} + \frac{s_2 D_0 + \beta_H N_0}{(s_2 - s_1)(s - s_2)(s + \beta_H)} \right\} \tag{23}$$

Using the inverse L. T. tables:

$$L - 1\left[\frac{1}{(s-a)(s-b)}\right] = \frac{1}{(a-b)}(e^{at} - e^{bt}) \tag{24}$$

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

$$H(t) = \lambda_D \left\{ \frac{s_1 D_0 + \beta_H N_0}{(s_1 - s_2)(s_1 + \beta_H)} e^{s_1 t} + \frac{s_2 D_0 + \beta_H N_0}{(s_2 - s_1)(s_2 + \beta_H)} e^{s_2 t} \right\} + \left\{ H_0 - \frac{\lambda_D(s_1 D_0 + \beta_H N_0)}{(s_1 - s_2)(s_1 + \beta_H)} - \frac{\lambda_D(s_2 D_0 + \beta_H N_0)}{(s_2 - s_1)(s_2 + \beta_H)} \right\} e^{-\beta_H t} \tag{25}$$

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

$$H(t) = \lambda_D \left\{ \frac{s_1 D_0 + \beta_H N_0}{(s_1 - s_2)(s_1 + \beta_H)} e^{s_1 t} + \frac{s_2 D_0 + \beta_H N_0}{(s_2 - s_1)(s_2 + \beta_H)} e^{s_2 t} \right\} \tag{26}$$

We should note here that Eqs (21) and (26) tend in 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 $s_1 = -\beta_H$ 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 $H(t) = 0$ 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'Hospital's Rule. Taking the limit of H(t) as $\lambda_D \to 0$ and hence $s_1 \to -\beta_H$, we get:

$$\lim_{\lambda_D \to 0} H(t) = \lim_{\substack{\lambda_D \to 0 \\ s_1 \to \beta_H}} \left\{ \frac{\lambda_D(s_1 D_0 + \beta_H N_0)}{(s_1 + \beta_D)(s_1 + \beta_H)} e^{-\beta_H t} + \frac{\lambda_D(-\beta_D D_0 + \beta_H N_0)}{(-\beta_D + \beta_H)(-\beta_D + \beta_H)} e^{-\beta_D t} \right\}$$

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 λ_D, as:

$$\lim_{\lambda_D \to 0} H(t) = \lim_{\lambda_D \to 0} \left\{ \frac{-\lambda_D \beta_H H_0 e^{-\beta_H t}}{(\beta_H - \beta_D)\{1/2[-(\beta_H + \beta_D + \lambda_D) - \sqrt{(\beta_H + \beta_D + \lambda_D)^2 - 4\beta_H \beta_D}] + \beta_H\}} \right\}$$

Differentiating the denominator and the numerator with respect to λ_D and then setting $\lambda_D = 0$ we get:

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

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

Expression for the deuterium 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:

$$R(t) = \frac{1}{\lambda_D} \left[\frac{(s_1 D_0 + \beta_H N_0)e^{s_1 t} - (s_2 D_0 + \beta_H N_0)e^{s_2 t}}{\frac{(s_1 D_0 + \beta_H N_0)}{(s_1 + \beta_H)} e^{s_1 t} - \frac{(s_2 D_0 + \beta_H N_0)}{(s_2 + \beta_H)} e^{s_2 t}} \right] \tag{27}$$

Dividing both the numerator and the denominator by N_0 and noting that $N_0 = H_0 + D_0$, we have:

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

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:

$$R(t) = \frac{1}{\lambda_D} \cdot \left[\frac{(s_1R_0 + \beta_H)e^{s_1t} - (s_2R_0 + \beta_H)e^{s_2t}}{\frac{(s_1R_0 + \beta_H)}{(s_1 + \beta_H)} e^{s_1t} - \frac{(s_2R_0 + \beta_H)}{(s_2 + \beta_H)} e^{s_2t}} \right] \tag{28}$$

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 state here that the D/H ratios observed in the Oklo phenomenon (which we denote as R_k) for Sample KN47 and Sample #556 were 131.2 and 126.1, respectively, even though those samples had water contents of 13.2% and 8.5%, respectively.[4]

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

$$R_k = \frac{1}{\lambda_D} \left[\frac{(s_1R_0 + \beta_H)e^{s_1t_d} - (s_2R_0 + \beta_H)e^{s_2t_d}}{\frac{(s_1R_0 + \beta_H)}{(s_1 + \beta_H)} e^{s_1t_d} - \frac{(s_2R_0 + \beta_H)}{(s_2 + \beta_H)} e^{s_2t_d}} \right] \tag{29}$$

where both s_1 and s_2 are functions of λ_D.

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

$$\lambda_D^{n+1} = F(R_k, \Phi, t_d, \lambda_D^n)$$

which implies that to obtain λ_D for a given R_k, Φ, and t_d, 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, Φ, the time duration t_d, and the deuteron disintegration constant, λ_D. The flux, $\phi = \Phi/t_d$, and β_H, β_D (the product of the cross section and the flux), and s_1, s_2 are functions of β_H, β_D, and λ_D, i.e., ϕ and λ_D:

Independent parameters:

Φ, t_d, λ_D

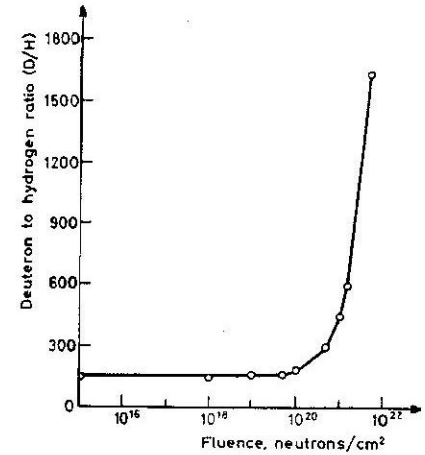

Fig. 2. D/H ratio for different fluences in case of no deuteron disappearance

Dependent parameters:

$\phi = \Phi/t_d$, $\beta_H(\phi)$, $\beta_D(\phi)$, $s_1(\phi, \lambda_D)$ $s_2(\phi, \lambda_D)$.

The fluence Φ and time duration t_d of the reaction that existed at Oklo are estimated based on the fission product concentrations, i.e., rate of production and subsequent decay. The fluence that is believed to have existed in Oklo, which explains the fission products isotopic changes,[1,3,4] is 10^{21} with slight variation from one location to another within the deposit. The duration of the reaction is estimated to have been between 600,000–1,200,000 years.[2]

Results

We calculated the value of the deuteron disintegration constant λ_D under the conditions of the Oklo phenomenon for three values of Φ 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: $3 \cdot 10^5$–$1.2 \cdot 10^6$ years. Although the variation of the estimated time duration is large, the value of λ_D 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 λ_D 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 $t_d = 10^6$ years.

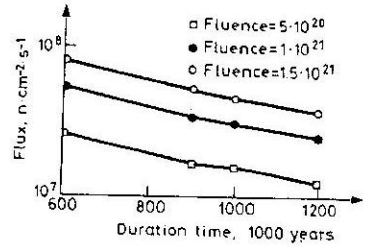

Fig. 3. Flux in the Oklo reaction for different fluences and time durations

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 $3.17 \cdot 10^7$ $n \cdot cm^{-2} \cdot s^{-1}$ which meets the condition that:

$$\phi << \lambda^{239}Pu/\sigma_\gamma^{239}Pu$$

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

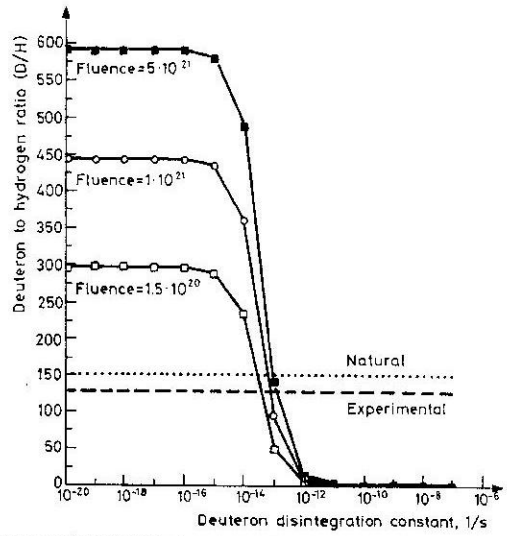

Fig. 4. Deuteron to hydrogen (D/H) ratio versus the deuteron disintegration constant for different fluences

between 10^{-20} and $10^{-6} \cdot s^{-1}$ for the three fluence cases for $t_d = 10^6$ years. The value of R at $\lambda_D = 10^{-20}$ ~ zero 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 λ_D less than 10^{-15}.

One can compute λ_{Dk}, i.e., the value of λ_D that will lead to a D/H ratio $R = R_k = 127.3$ (from the Sample Oklo #310) for a reaction in which the fluence was Φ and have lasted for time t_d. For the reference case, this turns out to be $7.47 \cdot 10^{-14} \cdot s^{-1}$.

Figure 5 shows the variation of R with λ_D for $\Phi = 10^{21}$ for different time durations. At $t_d = 6 \cdot 10^5$ y, $\lambda_D = 1.24 \cdot 10^{-13}$ and for $t_d = 10^6$, $\lambda_D = 7.47 \cdot 10^{-14} \cdot s^{-1}$. Figure 6 shows the variation of λ_{Dk} with the time period of the Oklo reaction for the three fluence cases.

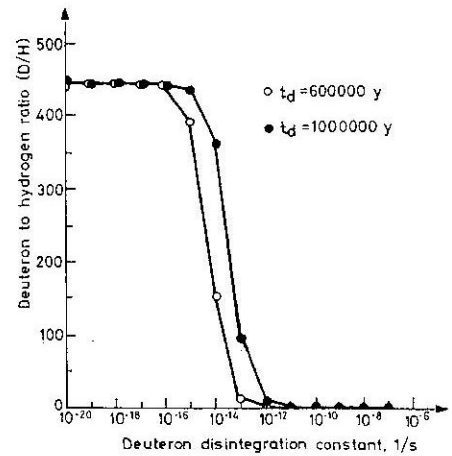

Fig. 5. Deuteron to hydrogen (D/H) ratio versus the deuteron disintegration constant for different reaction durations (t_d)

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:

$$_1D^2 + {}_ZN^A \rightarrow {}_1H^1 + {}_0n^1 + {}_ZN^A \qquad (30)$$

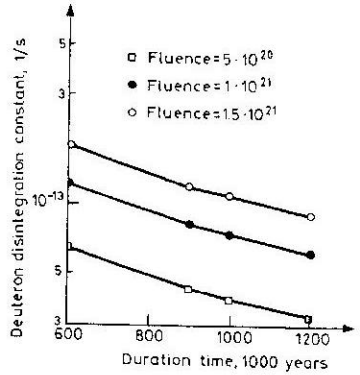

Fig. 6. Deuteron disintegration constant at Oklo versus reaction duration (t_d) for different fluences

here the nucleus $_ZN^A$ 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 $_ZN^A$ according to[5,6]:

$$_1D^2 + {}_ZN^A \rightarrow {}_1H^1 + {}_ZN^{A+1} \tag{31}$$

An example of this kind of reaction would be:

$$_1D^2 + {}_1D^2 \rightarrow {}_1T^3 + {}_1H^1$$

$$_1T^3 \rightarrow {}_{-1}e^0 + {}_2He^3$$

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:

$$_1D^2 + {}_1D^2 \rightarrow {}_{-1}e^0 + {}_1H^1 + {}_2He^3 \tag{32}$$

Compare this to the same reaction proceeding by neutron capture:

$$_1D^2 + {}_0n^1 \rightarrow {}_1T^3$$

$$_1T^3 \rightarrow {}_{-1}e^0 + {}_2He^3.$$

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

$$_1D^2 + {}_0n^1 \rightarrow {}_{-1}e^0 + {}_2He^3 \tag{33}$$

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 hyothesis can be tested based on the outcome of rections 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 $\sim 10^{-3}$ of the initial uranium concentration. Denoting the proportionality constant in Eq. (34) by μ_D, we obtain:

$$\lambda_D = \mu_D N$$

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

We note that μ_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:

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

However, the result is not valid for a D–D disintegration reaction as the term describing the deuteron disintegration in this case should be λD^2. We have accounted for this situation in the numerical calculation scheme and the D–D disintegration constant that will account for the D/H ratio R_k is found to be $\sim 10^{-31}$ 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].

Tritium and helium-3 concentrations

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

$$dT/dt = \beta_D D(t) - \lambda_T T + \beta_p He \tag{36}$$

$$dHe/dt = \lambda_T T - \beta_p He \tag{37}$$

We shall solve first for the case where ^3He does not escape from the site of the reaction and shall account for the ^3He (n, p) reaction. We shall obtain the case where ^3He leaks out of the site as a special case when $\beta_p = 0$. We shall show that the concentration of ^3He 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 ^3He are zero and ^3He does not leak out, we get:

$$T(t) = N_0\beta_D\left\{\frac{(s_1+\beta_p)(s_1R_0+\beta_H)}{s_1(s_1-s_2)(s_1-s_3)}e^{s_1t} + \frac{(s_2+\beta_p)(s_2R_0+\beta_H)}{s_2(s_2-s_1)(s_2-s_3)}e^{s_2t} + \frac{(s_3+\beta_p)(s_3R_0+\beta_H)}{s_3(s_3-s_1)(s_3-s_2)}e^{s_3t} - \frac{\beta_p}{\beta_D s_3}\right\} \tag{38}$$

and

$$He(t) = N_0\lambda_T\beta_D\left\{\frac{(s_1R_0+\beta_H)}{s_1(s_1-s_2)(s_1-s_3)}e^{s_1t} + \frac{(s_2R_0+\beta_H)}{s_2(s_2-s_1)(s_2-s_3)}e^{s_2t} + \frac{(s_3R_0+\beta_H)}{s_3(s_3-s_1)(s_3-s_2)}e^{s_3t} - \frac{1}{\beta_D s_3}\right\} \tag{39}$$

where λ_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 ^3He are not zero. However, there is no reason to assume that T or ^3He 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 ^3He are proportional to the initial water content in the rock. For the typical case of $\Phi = 10^{21}$, $t_d = 10^6$ so that $\lambda_D = 7.47 \cdot 10^{-14}$ the change in T and He concentrations with time is shown in Fig. 7. The final concentrations in this case are $T(t_d) = 3.8 \cdot 10^7$ and $He(t_d) = 3.78 \cdot 10^{11}$. The rate of ^3He production would be $He(t_d)/t_d = 0.012$ ^3He atom/cm$^3 \cdot$ s.

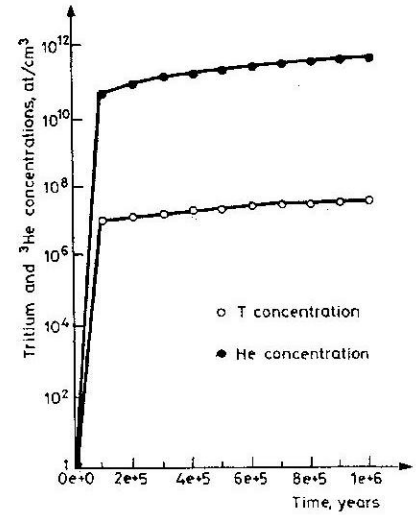

Fig. 7. Variation of tritium and helium-3 concentrations with time

If ^3He 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:

$$dT/dt = \beta_D D - \lambda_T T \tag{40}$$

$$dHe/dt = \lambda_T T \tag{41}$$

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 $3.78 \cdot 10^{11}$ at/cm^3. If all the helium-3 produced escapes, the rate of ^3He release to the atmosphere would be 0.012 at/cm$^3 \cdot$ s.

To explain why the ^3He concentration does not change when setting $\beta_p = 0$, we note that β_p appears in Eq. (33) for He within the term $s_3 = -(\beta_p + \lambda_T)$. For the Oklo cases, $\beta_p \sim 10^{-13}$ and $\lambda_T = 1.79 \cdot 10^{-9}$, so that $\lambda_T >> \beta_p$ and $s_3 = -\lambda_T$. Hence β_p will not affect the ^3He concentration.

Discussion

The deuteron has the following unique characteristics[5,6] compared with other nuclei:

(1) It is the only two-particle nuclear bound system.

(2) It has the lowest Binding Energy (B. E.) among other nuclei. Table 2 compares the B. E. of deuterons with those of other nuclei.

Table 2
Binding energy (BE) of typical nuclides[5]

Nuclide	BE/A, MeV/nucleon	BE, MeV
^{2}D	1.12	2.23
^{3}T	2.83	8.48
^{3}He	2.57	7.72
^{4}He	7.08	28.30
^{6}Li	5.33	32.00
^{7}Li	5.60	39.20
^{9}Be	6.47	58.19
Average per nucleon:	~8.50	—

(3) Deuterons do not possess any stable excited state.

(4) In the deuteron nucleus, the neutron and the proton spend about one half of the time outside the range of the nuclear forces. Using the equation $R = 1.25\ A^{1/3}$ fm, (1 fm = 10^{-13} cm), the radius of the deuteron will be 1.6 fm. The separation between the neutron and the proton in the deuteron nucleus is 4 fm.

(5) The change distribution of the deuteron nucleus is very unsymmetrical.

(6) The separation between the center of mass and the center of change in the deuteron nucleus is the most extreme among other nuclei.

These characteristics suggest that even though the deuteron is stable with respect to natural radioactive decay, it is a very loose particle that may break up if it existed in conditions that will further enhance the instabilities in its nuclear structure to the point of disintegration.

Having observed an anomaly in the $^{238}U/^{235}U$ ratio that would have implied that fast fissions of ^{238}U had occurred, VLASOV[7] suggests that the fast fissions in ^{238}U and the value of 0.5 for the regeneration ratio of ^{235}U can be explained by assuming that an annihilation explosion of an antimatter meteorite caused the formation of free fast neutrons. He further estimates the yield of such an explosion to be from 3–5 neutrons per antinucleon annihilation depending on the conditions. According to our hypothesis, the postulate of an antimatter explosion at the Oklo site is not necessary to

explain the anomalous $^{238}U/^{235}U$ ratio. According to the proposed model, ^{238}U can undergo the following reactions:

$$^{2}_{1}D + ^{238}_{92}U \rightarrow F_1 + F_2 + ^{1}_{1}H + \nu_0 n^1$$

or

$$^{2}_{1}D + ^{238}_{92}U \longrightarrow ^{1}_{1}H + ^{239}_{92}U$$

where F_1 and F_2 are fission products and ^{239}U eventually produces ^{239}Pu. The same two reactions are possible with ^{235}U. Deuteron disintegration would produce a source of neutrons S, the number of such neutrons will be given by $\lambda_{Dk}D(t)$. For the typical case, if we take the average value of the deuteron concentration to be $\sim (D_i + D_f)/2$, then the number of neutrons will be equal to $6.44 \cdot 10^4$ fast neutrons/(cm$^2 \cdot$ s). If we denote the multiplication factor of the assembly by k, then the flux is given by:

$$\phi = S + kS + k^2S + k^3S + k^4S + \ldots = S(1 + k + k^2 + k^3 + k^4 + \ldots)$$

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

$$\phi = S/(1 - k).$$

Since $\phi = 3.17 \cdot 10^7$, and $S = 6.44 \cdot 10^4$, thus $k = 0.998$, implying a subcritical, source-driven system.

JONES et al.[8] suggest that cold nuclear fusion takes place in the earth interior. Based on a ^3He flow out of the earth mantle of $2 \cdot 10^{19}$ ^3He atom/s, and that the mantle water reservoir is $1.4 \cdot 10^{24}$ g, they estimate a value for the D–D fusion constant of 10^{-24} fusion/deuteron · s. The value which we calculated for a D–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 ^3He flow out of the earth's mantle. We have calculated that under the localized conditions of the Oklo deposit, the rate of ^3He escape out of the deposit is 0.012 ^3He at/cm^3 · 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 $1.4 \cdot 10^{24}$ cm^3 then we can calculate the total ^3He flow out of the earth mantle due to a reaction similar to the Oklo reaction to be $1.7 \cdot 10^{22}$ ^3He at/s. This can be compared to JONE'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.[8]

Deuteron disintegration may be considered as a possible explanation of some

observation of deuterium reactions in palladium and titanium electrodes.[9] 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 butter understanding of these observations.

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