

## ANOMALOUS DEUTERON TO HYDROGEN RATIO IN OKLO SAMPLES AND THE POSSIBILITY OF DEUTERON DISINTEGRATION

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### Abstract

A hypothesis is presented to explain the anomalous D/H ratio observed in samples from the site of the naturally occurring fission reaction at Oklo. The experimentally observed D/H ratio of 127 ppm exceeds the naturally occurring value of 150 ppm. Further, using a multicomponent system consisting of hydrogen, deuterium, tritium and helium nuclei to model the Oklo reaction phenomenon and assuming a thermal fission process term, we calculate a D/H ratio of 445 ppm in the presence of the thermal neutron fluence attributed to Oklo. However, solving the same rate equations with a deuterium sink term to represent the hypothesis of deuteron disintegration, we find a deuteron disintegration constant of  $7.47 \times 10^{-14} \text{s}^{-1}$  yields the observed D/H ratio. Indeed, deuteron disintegration would provide a neutron source (in addition to the fission neutrons) that could have driven the Oklo system as a subcritical (vs. a critical) reactor over the extended period attributed to it.

## Introduction

The historic phenomenon where a naturally occurring fission chain-reaction took place is located at the Oklo mining site in Gabon, Africa.[1,2] Geological characteristics at Oklo allowed the formation of soluble uranium salts in water. Since fission products have been found in samples from the site, it was determined that a fission process had taken place in this ore. Based on the measured depletion of  $U^{235}$  and the concentrations of the fission products, the reaction is estimated to have lasted for a period between 600,000 to 3,500,000 years [3]. The fluence of neutrons required to account for these observations has been estimated by various investigators to be about  $10^{21}$  n/cm<sup>2</sup>[1-4].

## The D/H Anomaly

Samples from the Oklo site have been analyzed for water and hydrogen content by heating them to about 1200°C. The D/H ratio in natural water is close to 150 ppm. In contrast, when water from minerals at the Oklo site was analyzed, the D/H ratio was found to range around 127 ppm[5]. This discrepancy is even more serious when it is realized that, as we show later, for the neutron fluence estimated for Oklo, the D/H ratio should have increased to about 450 ppm because of neutron capture in hydrogen. To our knowledge this discrepancy, namely the low D/H ratio measured at Oklo, has not previously been discussed in the literature nor has any explanation been proposed prior to this.

## Analysis

Figure 1a depicts a multicomponent system that describes the major isotopic transitions for elements (Hydrogen, Deuterium, Tritium and He-3) undergoing a neutron irradiation. Assuming that all of the He<sup>3</sup> produced escapes

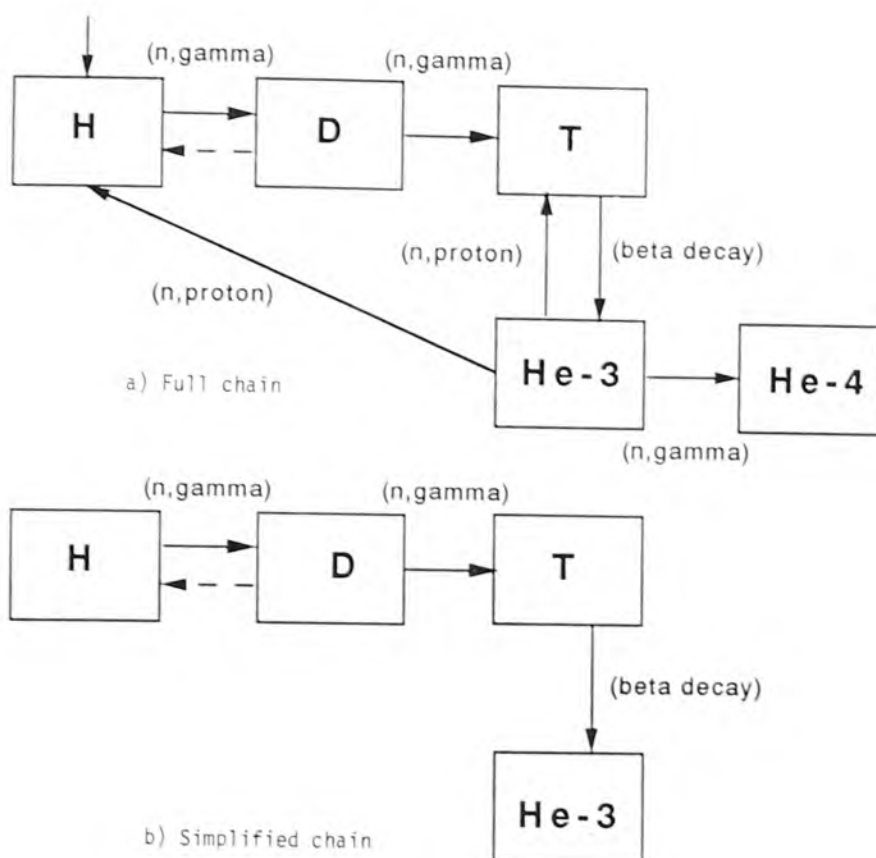


Figure 1. Multicomponent System Consisting of Hydrogen, Deuterium, Tritium, and Helium-3 and Isotopic Transitions of Elements in a Neutron Flux.

from the site so that it doesn't contribute further to the isotopic changes, the chain is simplified to the one shown in Fig. 1-b. (If  $\text{He}^3$  did not escape from the reaction site, it could either undergo an  $(n, p)$  or an  $(n, \gamma)$  reaction). Then, in this multicomponent system, hydrogen can be produced from two sources: 1) the  $\text{He}^3$   $(n, p)$  reaction, and 2) postulated deuteron disintegration according to:  ${}_1\text{D}^2 \rightarrow {}_0\text{n}^1 + {}_1\text{H}^1$ . Deuterons are produced from neutron capture in hydrogen and are provided with two sinks: disintegration into hydrogen [6,7], and capture of a neutron with production of tritium. Tritium decays by  $\beta^-$  emission to  $\text{He}^3$  with a half life of 12.3 years.

Because of the presence of water, we are justified to assume that the Oklo neutron energy spectrum was basically thermal with a Maxwellian shape at temperature  $T_0$ . Table I shows the thermal neutron cross sections for the various reactions in this chain.

The rate equations for the multicomponent system shown in Fig. 1, assuming a pure fission process but with the added hypothesis of deuteron disintegration (decay constant  $\lambda_D$ ) into  $H + n$ , can be written as:

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

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

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

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

Here  $H$ ,  $D$ ,  $T$ , and  $He$  are the concentrations in atoms/cm<sup>3</sup> of hydrogen, deuterium, tritium, and helium-3, respectively.

We have defined:

$$\beta_H = \sigma_{\gamma H} \phi, \quad (s^{-1}) \quad (5a)$$

$$\beta_D = \sigma_{\lambda D} \phi, \quad (s^{-1}) \quad (5b)$$

$$\beta_p = \sigma_{pHe} \phi \quad (s^{-1}) \quad (5c)$$

where we made the approximation:

$$\sigma_{pHe} \approx \sigma_{aHe} \quad (6)$$

**Table I. Thermal Cross Sections for H, D, T, and He<sup>3</sup> Neutron Reactions**

Reaction	Thermal Neutron Cross Section (Barns)
H (n, $\gamma$ ) D	0.333
D (n, $\gamma$ ) T	$0.51 \times 10^{-3}$
T (n, $\gamma$ ) H <sup>4</sup>	$< 6 \times 10^{-3}$
He <sup>3</sup> (n, p) T	5330
He <sup>3</sup> (n, $\gamma$ ) He <sup>4</sup>	0.05

This assumption implies that absorption in He-3 mainly cause the (n,p) reaction which has a large cross-section value as shown in Table I.

As a first step, we are interested in the H and D concentrations without deuterium disintegration ( $\lambda_D = 0$ ). Solving the corresponding set of coupled differential equations, we find the ratio  $R=D/H$  in this case is:

$$R = \left\{ R_0 + \frac{\sigma_{\gamma H}}{\sigma_{\gamma H} - \sigma_{\gamma D}} \right\} e^{(\sigma_{\gamma H} - \sigma_{\gamma D}) \phi} - \left( \frac{\sigma_{\gamma H}}{\sigma_{\gamma H} - \sigma_{\gamma D}} \right) \quad (7)$$

where the reference (starting) ratio is taken as the natural value, i.e.,  $R_0=D_0/H_0=150$  ppm.

We note that  $R$  is function of the fluence only, since all other parameters are constants. Using the estimated value of the fluence for Oklo,  $\phi = 10^{21}$  n/cm<sup>2</sup>, we find that  $R = 445$  ppm. This then is the  $R$  ratio expected under the natural thermal reactor assumption without deuteron disintegration.

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 the natural value  $R_0$  (150 ppm), when it should have risen to around 445 ppm as calculated above, is in direct contradiction with the expectations based on the natural reactor assumption alone. This anomaly is the focus of our study.

We next consider the case with postulated deuterium disintegration. This is a tempting assumption since, as seen from Table II, compared to other low-Z elements, deuterium has a uniquely low binding energy. Here we have assumed that the anomaly is due to a nuclear process since we are unaware of any chemical or other physical process

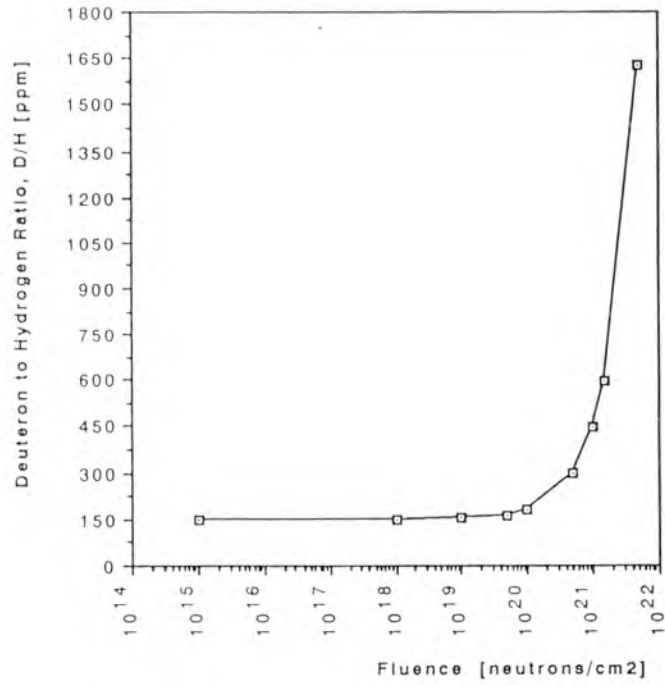


Figure 2. D/H Ratio for Different Fluences in Case of no Deuteron Disintegration ( $\lambda_D = 0$ ).

Table II. Binding Energy (BE) of Typical Nuclides [5]

Nuclide	BE/A (MeV/nucleon)	BE (MeV)
D-2	1.12	2.23
T-3	2.83	8.48
He-3	2.57	7.72
He-4	7.08	28.30
Li-6	5.33	32.00
Li-7	5.60	39.20
Be-9	6.47	58.19
Average per nucleon	~ 8.50	--

which could have depleted the deuterium and/or enriched the hydrogen content of the ore mineral. Thus we propose that the deuterons have undergone a process involving disintegration. Then, regardless of the mechanism involved, the following reaction would have, in principle, taken place:



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

$$R_k = \frac{1}{\lambda_D} \left[ \frac{(s_1 R_0 + \beta_H) e^{s_1 t_d} - (s_2 R_0 + \beta_H) e^{s_2 t_d}}{\frac{(s_1 R_0 + \beta_H)}{(s_1 + \beta_H)} e^{s_1 t_d} - \frac{(s_2 R_0 + \beta_H)}{(s_2 - \beta_H)} e^{s_2 t_d}} \right] \quad (9a)$$

where both  $s_1$  and  $s_2$  are functions of  $\lambda_D$ , i.e.

$$s_1, s_2 = 1/2 \{ -(\beta_H + \beta_D + \gamma_D) \pm [(\beta_H + \beta_D + \gamma_D)^2 - 4\beta_H \beta_D]^{1/2} \} \quad (9b)$$

### Results

We have calculated the value of the deuteron disintegration constant  $\lambda_D$  under the conditions of the Oklo phenomenon for three values of  $\Phi$  around  $10^{21}$ , namely:  $0.5 \times 10^{21}$ ,  $1.0 \times 10^{21}$ ,  $1.5 \times 10^{21}$ , and for different time durations in the range:  $3 \times 10^5$ – $1.2 \times 10^6$  years. Although the variation of the estimated time duration is large, the value of  $\lambda_D$  calculated for the same fluence and limiting time periods is not very large. Similarly, for the same time duration, the variation of  $\lambda_D$  for fluence values of  $0.5 \times 10^{21}$  and  $1.5 \times 10^{21}$  is also not large. Thus, we adopt as a reference case values of  $\Phi = 10^{21}$  and  $t_d = 10^6$  years. In Fig. 3 we show the D/H ratio,  $R$ , for the range of  $\lambda_D$  values between  $10^{-20}$  and  $10^{-6} \text{ s}^{-1}$  for the three fluence cases for  $t_d = 10^6$  years.



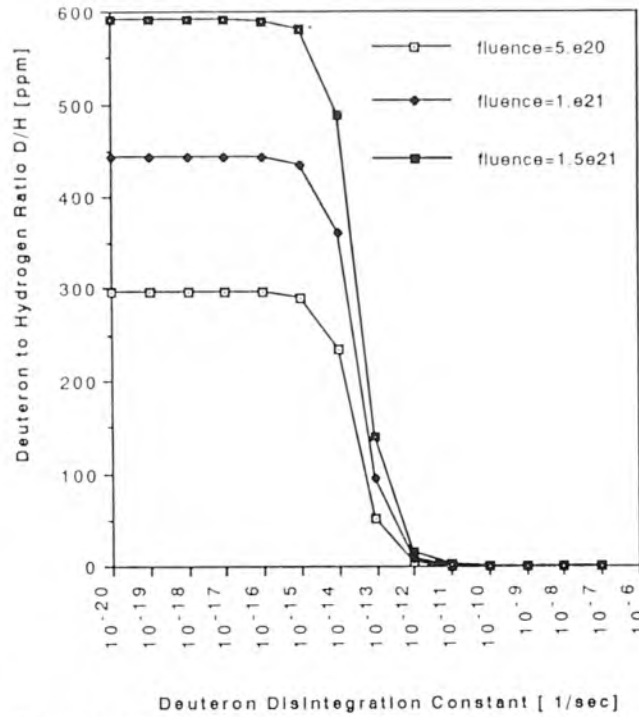
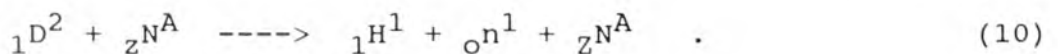


Figure 3. Deuteron to Hydrogen (D/H) Ratio Versus the Deuteron Disintegration Constant for Different Fluences.

One can compute  $\lambda_{Dk}$ , i.e., the value of  $\lambda_D$  that will lead to a D/H ratio  $R=R_k=127.3$  (from the Oklo sample #310) for a reaction where the fluence  $\Phi$  has lasted for time  $t_d$ . For the reference case, this turns out to be  $7.47 \times 10^{-14} \text{ s}^{-1}$ .

#### COMMENTS ABOUT PROCESSES

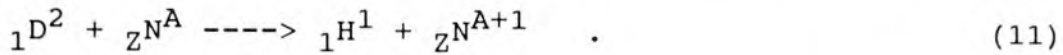
Up to this point the results are independent of the specific deuteron disintegration process assumed. Now it is interesting to consider several possible processes and the consequences implied. We assume that deuteron disintegration is not a single particle reaction but rather a multiple particle interaction, i.e., unlike natural decay, the deuteron disintegrates under the unique system created by the collection of the deuteron itself and the surrounding atoms under the thermodynamic conditions present. Then the deuteron disintegration reaction may proceed in one of two ways. First, postulating a free neutron reaction, we have:





Here the nucleus  ${}_Z\text{N}^A$  acts as a catalyst for the deuteron disintegration process, and does not undergo any isotopic changes.

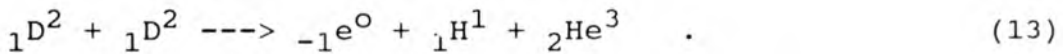
A second way for the reaction to proceed involves isotopic changes in the medium that involve a "neutron swapping" type reaction[6,7]. Then the deuteron gives up its neutron to the neighboring nucleus  ${}_Z\text{N}^A$  according to:



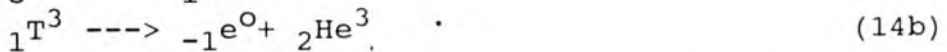
An example of this kind of reaction for Oklo would be:



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



It is instructive to compare this result to the deuterium transformation driven by neutron capture:



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



Both reactions involve the depletion of deuterons with the production of tritium and  $\text{He}^3$ . 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 neutron swapping hypothesis could be tested based on reactions 12-13 vs. 14-15.

According to the preceding disintegration assumption, we can state, in general, that the rate of disintegration of deuterons is proportional to the product of the deuteron concentration and to the concentration of host atoms  $N$ , i.e.:

$$\lambda_D D \propto D N \quad . \quad (16)$$

We further assume that the concentration of the host atom, corresponding to uranium in the Oklo case, is constant. This assumption is reasonable since the fractional uranium depletion in the estimated fluence will be only  $\sim 10^{-3}$ . Then denoting the proportionality constant in Eqn. 16 by  $\mu_D$ , we obtain:

$$\lambda_D = \mu_D N \quad (17a)$$

or

$$\mu_D = \lambda_D / N \quad \text{cm}^3/\text{s} \quad . \quad (17b)$$

Note that  $\mu_D$  has the same units as  $\langle \sigma v \rangle$ , the reaction parameter used in plasma physics and fusion studies.

If the reaction is a D-U disintegration reaction, i.e., deuterons disintegrating while present in the Uranium lattice, assuming that the mineral was in the form of  $\text{UO}_2$ , we find for the Oklo sample #310 data:

$$\mu_{DU} = \lambda_D / U = 2.24 \times 10^{-35} \text{ cm}^3/\text{s} \quad (18)$$

where a  $U$  concentration of  $3.33 \times 10^{21} \text{ U atoms/cm}^3$  was used.

The above result is not valid for D-D disintegration, however, since then the term describing deuteron

disintegration rate should be  $\lambda D^2$ . We have accounted for that situation in the numerical calculation scheme. Then the D-D disintegration constant needed to account for the D/H ratio  $R_k$  is found to be  $\sim 10^{-31} \text{cm}^3/\text{s}$ . Further, if the reaction involved was a D-D disintegration such that it can be described by the term  $\lambda_D D$ , then the reaction constant is  $\sim 7.47 \times 10^{-14}$  [D disintegrations/D.sec] as noted earlier.

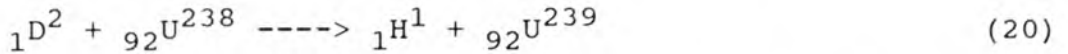
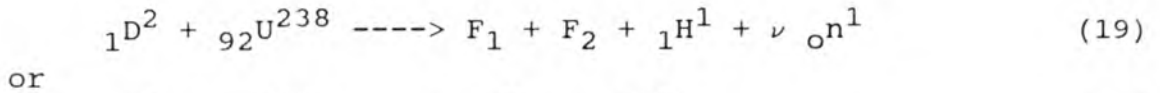
### **Tritium and Helium-3 Concentrations**

As shown in Fig. 1, the Oklo reaction will lead to the production of T and  $\text{He}^3$ . To obtain their concentrations, the rate Equations 3 and 4 are solved with  $D(t)$  given by the solution of Equations 1 & 2. It is possible to obtain a solution in the case where the initial concentrations of T and  $\text{He}^3$  are not zero. However, there is no reason to assume that T or  $\text{He}^3$  existed in the ore initially, and hence we will not show the solution in that case. Interestingly, an examination of these equations shows that the concentrations of T and  $\text{He}^3$  are proportional to the initial water content in the rock. For the typical case of  $\phi = 10^{21}$ ,  $t_d = 10^6$  so that  $\lambda_D = 7.47 \times 10^{-14}$  and the final concentrations in this case are  $T(t_d) = 3.8 \times 10^7$  and  $\text{He}(t_d) = 3.78 \times 10^{11}$ . The corresponding rate of  $\text{He}^3$  production would be  $\text{He}(t_d)/t_d = 0.012 \text{He}^3 \text{atom/cm}^3 \cdot \text{s}$ .

### **Uranium Anomaly**

Earlier Vlasov[8] pointed out that the  $\text{U}^{238}/\text{U}^{235}$  ratio was anomalously low. This seems to imply that fast neutron fission of  $\text{U}^{238}$  occurred. Thus, Vlasov suggests that an annihilation explosion of an antimatter meteorite caused the formation of free fast neutrons[8]. He further estimates the yield of such an explosion to be from 3-5 neutrons per antinucleon annihilation depending on the conditions. However, if our deuterium disintegration hypothesis is employed under the assumption of neutron swapping, the postulate of an antimatter explosion is not necessary. This

can be understood by considering the various reactions that may have occurred. According to the proposed neutron swapping model,  $U^{238}$  can undergo the following reactions:



where  $F_1$  and  $F_2$  are fission products. The  $U^{239}$  decay chain eventually produces  $Pu^{239}$  which could undergo thermal fission. This process could possibly explain the low  $U^{238}/U^{235}$  ratio.

#### COMPARISON WITH EARTH CORE HYPOTHESIS

Jones et al.[9] have suggested that cold fusion takes place in the earth interior. Based on an estimated  $He^3$  flow of  $2 \times 10^{19}$   $He^3$  atom/s out of the earth's mantle, and using a mantle water reservoir of  $1.4 \times 10^{24}$  g, they estimate a value for the cold fusion D-D fusion constant of  $10^{-24}$  fusions/deuteron-s. The value which we calculated for a D-D disintegration reaction in the Oklo medium is  $7.57 \times 10^{-14}$  disintegrations/deuteron-sec, which is about ten orders of magnitude higher than the Jones, et al. value.

We can estimate, according to our hypothesis, the  $He^3$  flow out of the earth's mantle. We have calculated that under the localized conditions of the Oklo deposit, the rate of  $He^3$  escape out of the deposit is  $0.012$   $He^3$  at/cm<sup>3</sup>.sec. If we assume that the same conditions that existed at Oklo prevail in the mantle of the earth, then we find a total  $He^3$  flow out of the earth's mantle due to an Oklo-type reaction is  $\sim 1.7 \times 10^{22}$   $He^3$  atm/sec. This is to be compared to the Jones, et al. value of  $2 \times 10^{19}$ , keeping in mind that the conditions of the Oklo reaction may have been more favorable than the earth's mantle for the deuteron disintegration reaction.

### Conclusion

The anomalously low D/H ratio found in samples from Oklo strongly suggest that a deuterium disintegration process occurred. If so, for Oklo conditions, the rate is shown to be about  $7.5 \times 10^{-14}$  D disintegrations/deuteron-sec. The exact mechanism involved is open to debate and study. We favor a neutron swapping type reaction which would be enabled by the unique Oklo conditions of thermodynamic parameters and D-U mixture. If so, this reaction could offer an alternate explanation to the anomalously low  $U^{238}/U^{235}$  ratio cited earlier by Vlasov. While the results show higher rates than those found by Jones, et al., for earth mantle reactions, the difference could be due to the specific conditions present at Oklo, e.g., the relatively high uranium concentration.

### Acknowledgments

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