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Limits on the variability of coupling constants from the Oklo natural reactor

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Thermal neutron capture cross sections in fission fragments need to be known to great precision in the nuclear power industry. These cross sections are frequently dominated by extremely narrow neutron capture resonances. An analysis of isotopic abundances at the site of a prehistoric natural reactor at Oklo in West Africa suggests that such a resonance in samarium-149 has moved by less than 0.01 eV in the past 2×10^9 years and this is used to place limits on the variability of coupling constants over this period.

1. Introduction

Typical nuclear interaction energies are measured in mega electron volts while thermal neutron capture cross sections in heavy nuclei are frequently dominated by extremely narrow resonances known to a precision of milli electron volts. These resonances thus provide a one part in 10^9 probe of the single-particle couplings in nuclei.

The discovery of the site of a prehistoric natural nuclear reactor at Oklo in Gabon, West Africa which was critical some 2×10^9 years ago provides us with geological samples, the isotopic abundances in which can be used to extract information on the operational characteristics of the reactor. Among these are thermal neutron capture cross sections. Limits on the variability of these cross sections place restrictions on the variability of the energies of single-neutron capture-resonances from which we may deduce limits on the variability of particle coupling strengths.

2. Reactor physics

The primary source of energy in a thermal nuclear reactor is the induced neutron fission of ^{235}U which is accompanied by the release of 2–3 neutrons,

$$\mathrm{n} + {}^{235}\mathrm{U} \rightarrow \text{fission fragments} + 2\text{–}3 \text{ neutrons}. \qquad (1)$$

When conditions are right for some of these neutrons released in fission to induce further fission then we have the possibility of a self-sustaining chain reaction.

The first problem that we encounter is that the neutrons released in fission have energies typically in the range 1–2 MeV while the neutron's effectiveness in inducing fission increases with the length of time it spends in the nucleus, that is, the lower its velocity and hence its energy. Neutrons in equilibrium with their surroundings will have a thermal distribution and in a reactor core at a temperature of $T \approx 1000\,\mathrm{K}$; this implies a typical energy $kT \approx 86\,\mathrm{meV}$. There is thus the need for a moderator to slow down the fission neutrons to thermal energies where they can effectively induce further fission. The most effective moderators are protons, and in the form of hydrogen these are readily available in water. A slight concern is that, besides slowing down neutrons, protons have a thermal neutron capture cross section of $0.33 \times 10^{-24}\,\mathrm{cm}^2$ which removes

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neutrons from the system so that they are no longer available to induce fission in the ^{235}U. This can be overcome by using deuterons in heavy water, rather than protons, as a moderator.

The second problem is the low relative abundance of ^{235}U which reduces the probability that a thermal neutron will encounter such a nucleus before it leaves the system or is captured by some other nucleus. The world-average ^{235}U isotopic abundance is 0.7202 ± 0.00014 %, the remainder being ^{238}U which is not amenable to thermal neutron induced fission. To overcome this problem uranium fuel for man-made reactors is artificially enriched, that is, the relative abundance of ^{235}U is increased. Clearly a light-water moderated reactor requires a higher level of enrichment than does one moderated by heavy water.

3. Fossil reactors

(*a*) *General considerations*

The relative shortage of ^{235}U can be traced to the half-lives of the uranium isotopes which are 7×10^8 years and 5×10^9 years for ^{235}U and ^{238}U respectively. Thus in the past there was more uranium than there is today and a higher proportion of it was ^{235}U. The half-lives of all other uranium isotopes are so much shorter that their natural abundances have been negligible throughout geological times. In table 1 we present the natural abundance of ^{235}U in the past based upon the present day level of 0.72 %. For comparison the degree of uranium enrichment required by man-made light-water reactors is 3–5 %. Thus we can see that it is extremely unlikely that an accidental accumulation of uranium-rich ore and water could have gone critical in the last 1.5×10^9 years.

Table 1. The past enrichment of uranium ores

age ($10^6 \times$ years) ...	0	700	1400	2100	2800
percentage ^{235}U	0.72	1.3	2.3	4.0	7.0

Besides the necessary level of enrichment, an additional requirement before a natural nuclear reactor could go critical would be the concentration of a sufficient mass of uranium. Impurities can migrate through crystalline materials ending up concentrated along fault lines and planes producing thin veins of mineral deposits. The chances of an essentially one- or two-dimensional reactor core achieving criticality are negligible. The development of a truly three-dimensional ore rich abundance requires mass transport and deposition of the element concerned. This is most easily achieved if the material is water soluble. In the case of uranium this means the existence of uranium oxides. It is generally believed that atmospheric oxygen first became available through photosynthesis some 2×10^9 years ago. Thus conditions suitable to the formation of a natural nuclear reactor seem limited to a period $1.5–2 \times 10^9$ years ago.

(*b*) *Oklo*

In 1972 the French C.E.A., monitoring uranium ores for their nuclear fuel enrichment programme, discovered samples that were heavily depleted in ^{235}U. Samples as low as 0.2 % were found and those with 0.4 % ^{235}U were typical. These ores came from the Oklo mines in Gabon, West Africa. A study of the site has led to the interpretation that the ^{235}U depletion is due to burn up in a natural reactor core.

The present-day characteristics of the Oklo ore bodies are consistent with conditions 1.8×10^9 years ago being similar to those in a man-made p.w.r. They suggest that criticality was achieved

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$1.84 \pm 0.07 \times 10^9$ years ago, that the reactor was operational for $2.29 \pm 0.7 \times 10^5$ years (although whether the operation was pulsed or continuous is not clear) and that integrated neutron fluxes of $1\text{–}2 \times 10^{21}$ neutrons cm^{-2} were experienced.

In support of these conclusions we present an analysis of neodymium isotopic abundances in table 2. In columns 1 and 2 of this table we show the isotopic abundances of two samples from Oklo. While there is generally a good correlation between the two samples there is an exception in the case of ^{142}Nd which is almost four times more prevalent in sample 2. Neither of the Oklo samples show any similarity to the world-average abundances for these isotopes presented in column 3. The only one of these isotopes not produced as a fission product is ^{142}Nd. We can use the ^{142}Nd abundances and the 'natural' abundances from column 3 to obtain the 'abnormal' abundances for the Oklo samples given in columns 4 and 5. The two samples now show an enhanced correlation and a striking resemblance to the ^{235}U fission yields presented in column 6.

TABLE 2. NEODYMIUM ABUNDANCES (%)

	Oklo 1	Oklo 2	world average	Oklo 1*	Oklo 2*	$^{235}U_f$
^{142}Nd	1.38	5.49	27.11	0	0	0
^{143}Nd	22.1	23.0	12.17	22.6	25.7	28.8
^{144}Nd	32.0	28.2	23.85	32.4	29.3	26.5
^{145}Nd	17.5	16.3	8.30	18.05	18.4	18.9
^{146}Nd	15.6	15.4	17.22	15.55	14.9	14.4
^{148}Nd	8.01	7.70	5.73	8.13	8.20	8.26
^{150}Nd	3.40	3.90	5.26	3.28	3.46	3.12

The only discrepancy between the samples and the fission yields is the slightly low readings for the odd isotopes ^{143}Nd, ^{145}Nd and the slightly high readings for the even isotopes ^{144}Nd, ^{146}Nd in the samples. This is to be expected if the ore has been exposed to a neutron flux in the reactor core, since the odd neodymium isotopes have substantial thermal neutron capture cross sections for the formation of the even neodymium isotopes. This is clarified in table 3 where we have added together the ^{143}Nd and ^{144}Nd and the ^{145}Nd and ^{146}Nd abundances to yield an enhanced agreement between the averaged Oklo 'abnormal' abundances and those resulting from ^{235}U fission. The relative ^{143}Nd to ^{144}Nd and ^{145}Nd to ^{146}Nd abundances are consistent with exposure to an integrated thermal neutron flux of 10^{21} neutrons cm^{-2}.

TABLE 3. NEODYMIUM ABUNDANCES (%)

	143 + 144	145 + 146	148	150
world average	36.02	25.52	5.73	5.62
$^{235}U_f$	55.18	33.53	8.16	3.13
Oklo	54.95	33.49	8.25	3.34
95% U_f + 5% Pu_f	54.78	33.65	8.28	3.29

In any nuclear reactor some fast neutrons will be captured by ^{238}U nuclei before they are moderated. The resulting ^{239}U can beta-decay to form ^{239}Pu

$$n + {}^{238}U \rightarrow {}^{239}U \underset{\beta^-}{\rightarrow} {}^{239}Np \underset{\beta^-}{\rightarrow} {}^{239}Pu, \qquad (2)$$

which is susceptible to thermal neutron induced fission. The fission yields from ^{239}Pu are slightly different from those of ^{235}U. A best fit to the Oklo abundances is given by a 5% contamination by ^{239}Pu fission yields (see the bottom row of table 3). The relative plutonium to uranium fission

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yields is a sensitive measure of the moderator to uranium content of the reactor core and suggests for the Oklo reactor a water to uranium abundance of 15 %.

(*c*) *The samarium isotopes*

In table 4 we present an analysis of the isotopic abundances for the samarium isotopes. The results are reminiscent of those for the neodymium isotopes. The Oklo abundances are very different from the world-averages; ^{144}Sm is not a fission product and can be used to determine the 'abnormal' Oklo samarium abundances; The abnormal samarium abundances are reminiscent of the ^{235}U fission yields; In the Oklo samples the odd isotopes show a depletion relative to the even isotopes consistent with an integrated neutron flux of 10^{21} neutrons cm^{-2}. The difference between the samarium and neodymium analysis is the extreme depletion of ^{149}Sm due to enormous thermal neutron capture cross section of 4.2×10^{-20} cm^2. This can be compared with the thermal neutron capture cross sections for ^{143}Nd, ^{145}Nd and ^{147}Sm which are 3.3×10^{-22} cm^2, 5×10^{-24} cm^2 and 9×10^{-23} cm^2 respectively.

Table 4. Samarium abundances (%)

	Oklo	world average		Oklo*	$^{235}U_f$
^{144}Sm	1.2	3.2		0	0
^{147}Sm	40.0	15.0	26.3	61.2	61.6
^{148}Sm	5.3	11.3			
^{149}Sm	3.9	13.9	31.4	27.3	29.3
^{150}Sm	21.4	17.5			
^{152}Sm	17.5	26.7		10.5	7.2
^{154}Sm	10.7	22.5		1.8	1.9

With the integrated neutron flux fixed at 10^{21} neutrons cm^{-2} the ^{149}Sm/^{147}Sm abundance ratio requires that the ^{149}Sm thermal neutron capture cross section during the operation of the Oklo reactor must have been within 10 % of its present day value.

Details of the findings at the Oklo mines are presented in an I.A.E.A. 1975 symposium proceedings.

4. Conclusions

The extraordinarily large ^{149}Sm thermal neutron capture cross section is due to the existence of a well established neutron resonance at 98 meV of width 63 meV which is firmly spanned by the thermal neutron distribution. In contrast the corresponding resonance in ^{147}Sm is at 3.4 eV and is well outside the thermal régime.

The total thermal neutron capture cross section in ^{149}Sm is the result of enfolding the energy dependent neutron capture cross section with the thermal distribution function. The thermal distribution is peaked at $\frac{1}{2}kT$ and has a width of order kT (remember $T \approx 1000$ K implies $kT \approx 86$ meV) whereas the neutron capture cross section is dominated by an extremely narrow resonance of width 63 meV. The total thermal neutron capture cross section is thus extremely sensitive to the relative position of these two highly peaked functions. Today's value of 4.2×10^{-20} cm^2 is a measure of the proximity of the resonance at 98 meV to the thermal peak. In order that the cross section was within 10 % of today's value during the operation of the Oklo reactor this neutron resonance must have been within 0.01 eV of its present day position.

The interaction of a neutron with a rare earth nucleus like ^{149}Sm is well described by an optical potential *ca.* 50 MeV deep and hence the limitation on the variation of the neutron resonance to

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less than 0.01 eV translates into a limit on the variation of the neutron coupling to the nucleus of 2×10^{-10} over the past 2×10^9 years or less than one part in 10^{19} per year over this period. Since this coupling comes *ca.* 95 % from the strong interactions, *ca.* 5 % from electromagnetic effects and *ca.* 10^{-5} % from the weak interactions, we deduce that the corresponding coupling constants have altered by less than one part in 10^{19}, 5×10^{17} and 10^{12} per year respectively over the past 2×10^9 years.

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