Title: Time variation of fundamental constants and the Oklo phenomenon

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Abstract:

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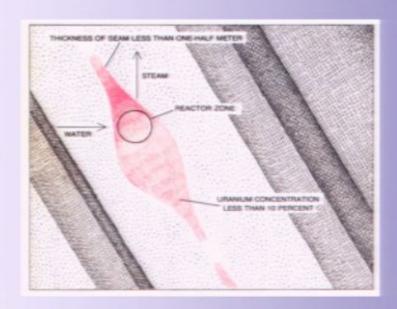
Time variation of fundamental constants and the Oklo phenomenon

Justin R. Torgerson

Los Alamos National Laboratory

Steven K. Lamoreaux

Yale University



- Site of several naturally occurring uranium fission reactors
- Effects of natural reactors discovered in 1972



Discovery of Oklo Phenomenon

- H. Bouzigues, on the staff of the Pierrelatte fuel processing plant in France, finds during routine analysis a sample that is anomalously low in U-235, 0.7171% compared to the usual 0.7202% in natural uranium ore. (May 1972)
- With rare exceptions (Pb, Os, He) the natural isotopic ratios of elements are remarkably constant.
- UF₆ gas used in mass spectroscopy might have been contaminated with spent fuel; that possibility was ruled out.
- Other explanations included, but not limited to:
 - Crash of nuclear powered spacecraft
 - Prehistoric civilization
 - U-235 somehow stolen
- The anomalous samples were traced to ore from a very concentrated uranium deposit in Gabon, Africa. (Oklo Deposit)
- Some samples showed depletion of U-235 to a level of 0.35%



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Explanation of low ²³⁵U abundance

Paul Kazuo Kuroda (1956, Univ. of Arkansas), in two scholarly papers, discussed the stability of uranium ore deposits. With the proper conditions, in the distant past, a uranium ore deposit could become "critical" and operate as a nuclear reactor.

P.K, "On the nuclear physical stability of the uranium minerals," J. Chem. Phys. 25, 781 (1956) P.K. "On the infinite multiplication constant and the age of uranium minerals," ibid., p. 1295 George A. Cowan, Sci. Am., p. 35, July 1976



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Requirements for a natural reactor

Relatively high U-235 isotopic concentration

The reason why the natural reactor was possible 2 billion years ago is given by the following data. The essential fact is that the half life of ²³⁵U is smaller than that of ²³⁸U.

Nat. Abund. (Present)
$$0.720\%$$
 99.27% Half Life $7\times 10^8 \text{years}$ $4.5\times 10^9 \text{years}$ Nat. Abund. $(2\times 10^9 \text{ years ago})$ 3.7% 96.3%

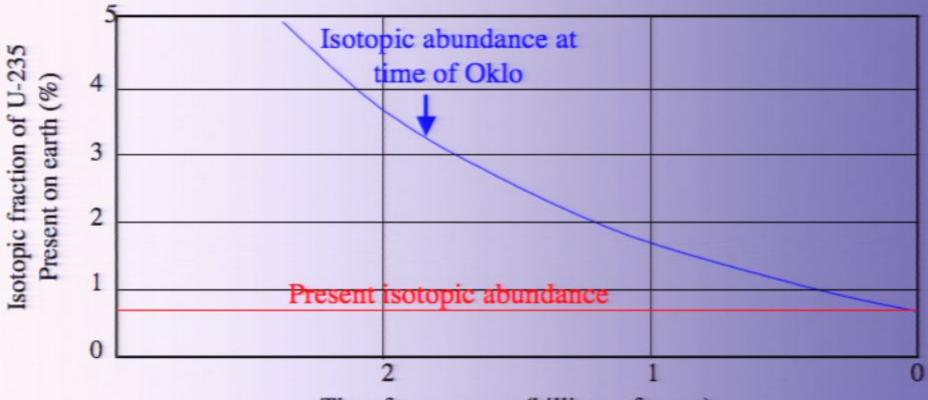
Based on these facts, K. Kuroda first predicted [7] that with the presence of some amount of water, the vein of uranium could undergo a chin reaction spontaneously. In 1972, the uranium slightly depleted in ²³⁵U was detected at French uranium-enrichment plant. It was traced and found out that at Oklo in Gabon Republic, uranium with ²³⁵U abundance of 0.4% - 0.5% was mined. It is well proved now that in 2 billion years ago, the reactors were operating in Oklo area (Oklo, Oklobondo, Bagombe). In spite of serious efforts to find the natural reactors in other area, no evidence was found up to now.

- Low concentration of neutron absorbers
- Presence of water to provide moderation
- Sufficiently large deposit





Isotopic fraction of U-235 through time



- Time from present (billions of years)
- U-235 decays about 6x faster than U-238
- Isotopic abundance of U-235 was 3.2% 1.8 billion years ago (25% when the earth formed 4.6 billion years ago)
- Isotopic abundance should be the same everywhere in the solar system





Some conditions for a homogeneous reactor

TABLE 18.1

VALUES OF p, f, AND k AND RATIO OF URANIUM TO MODERATOR AT MAXIMUM k FOR VARIOUS UNIFORM MIXTURES OF MODERATOR AND NATURAL URANIUM

Moderator	Molecules of Moderator Atoms of Uranium	f	p	k
H ₂ O	2.43	0.828	0.755	0.838
D ₂ O (+0.2% H ₂ O)	167	.955	.910	1.145 0.796
Be	193 452	0.837	0.757	0.849

k = 1.38 for 3.7% U-235 and 2.4 H₂0/U

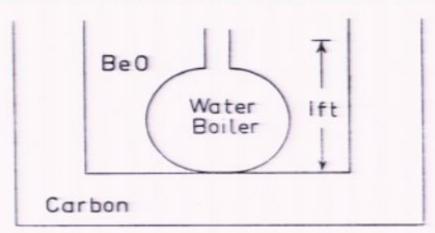


Fig. 83. - Los Alamos water boiler.

Possibility for a test of time variation of fundamental constants using Oklo

- Isotope with low-energy neutron absorption resonance (eg. ¹⁴⁹Sm, ¹⁵⁵Gd, ¹⁵⁷Gd)
- Accurate data for initial and final isotope abundance
 - Isolated, preserved samples
 - Natural abundance
 - Reactor-produced abundance (fission products, neutron captures, etc.)
 - Effect of neutron captures
- Energy-dependence of neutron resonance
 - Energy-dependence of other parameters such as decay rates (usually assumed to be independent of energy to first order)
- Model for Oklo reactor dynamics
- Model for relating change in resonance energy to change in fundamental constant(s)





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History of searches for time variation of α with Oklo

Basic idea due to A.I. Shlyakhter, Nature 264, 340 (1976) $\left|\frac{\dot{\alpha}}{\alpha}\right| < 1 \times 10^{-17}/yr$

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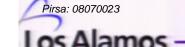
Lamoreaux and Torgerson use improved moderated neutron spectrum, Phys. Rev. D 69, 121701(R) (2004)

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 $\frac{\dot{\alpha}}{\alpha} = + \left(2 + 6 \atop -5\right) \times 10^{-18} / yr$

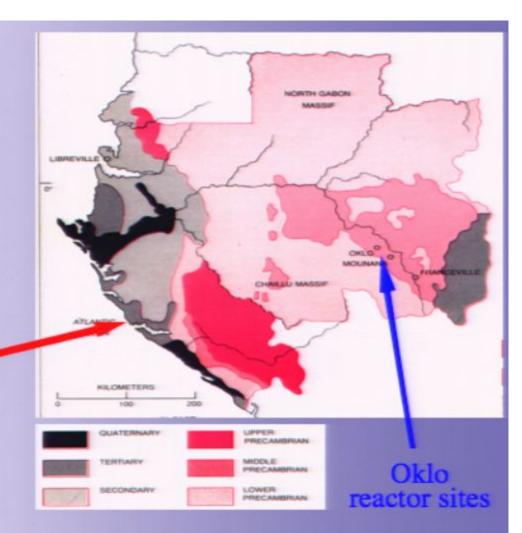
$$\frac{d\ln(E_r)}{dt} = c_{\alpha} \frac{d\ln(\alpha)}{dt} + c_{\Lambda} \frac{d\ln(m_q/\Lambda_{QCD})}{dt}$$





Where is Oklo?





- Most of Gabon is over 600M years old
- Mine in sedimentary rock bordered by older igneous rock





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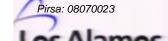
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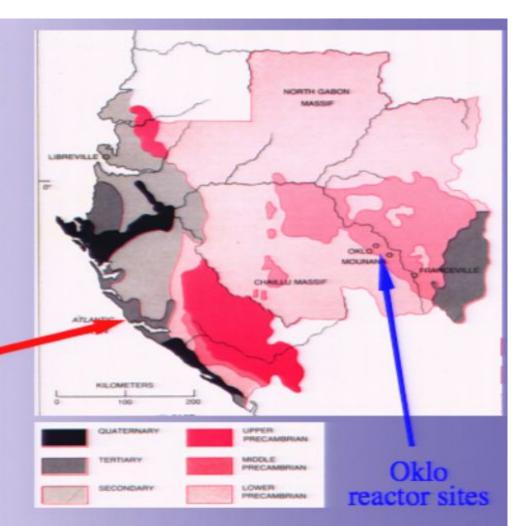
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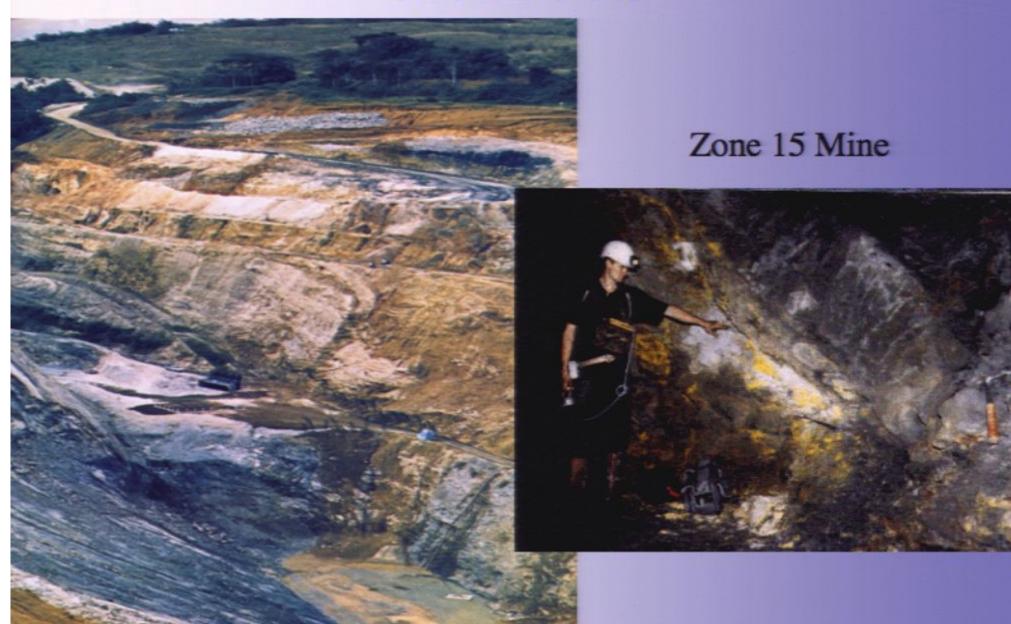


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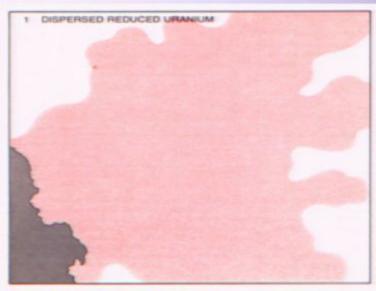




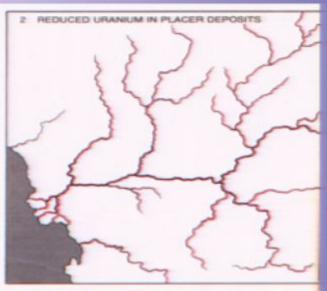
Oklo mine site



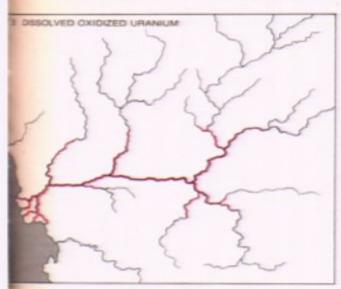
Formation of ancient uranium deposits



ACCRETION OF THE ORE DEPOSIT at Oklo involved the concentration of uranium derived from an entire watershed. The uranium (color) was originally dispersed in igneous rocks over a large area (1). As the rocks were eroded by weathering, the uranium accumulated in streambeds in a



relatively reduced state (2). It may have formed placer deposits like those in which gold and low-grade, Pre-cambrian arantum over are frund today. Some two billion years ago a biological development profoundly altered the disposition of the uranium.



the proliferation of photosynthetic algae increased the concentration of oxygen in the water. Highly oxidized uranium is more while than the reduced form, and the uranium was therefore dissolved and carried downstream (3). It remained in solution

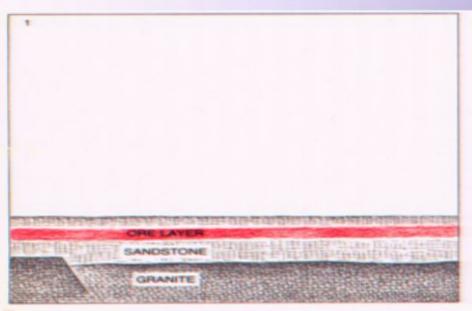


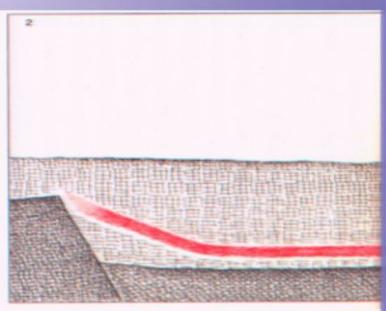
until it reached the delta of the river system, where sediments rich in organic noze again created a condition of oxygen deficiency. In these sediments the uranium was returned to the reduced form and was precipitated out of the solution (4). In the delta the ore was covered by later sediments.

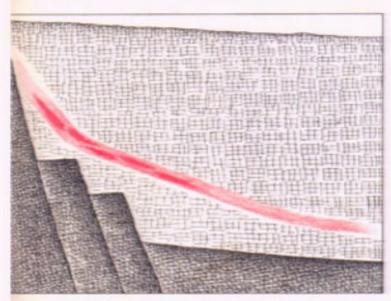


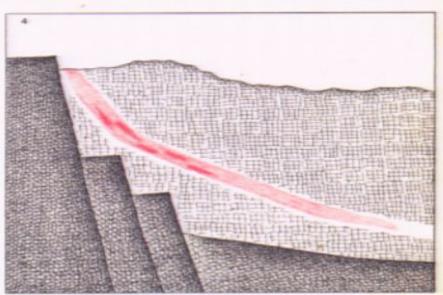
Physics

Evolution of the ore deposit

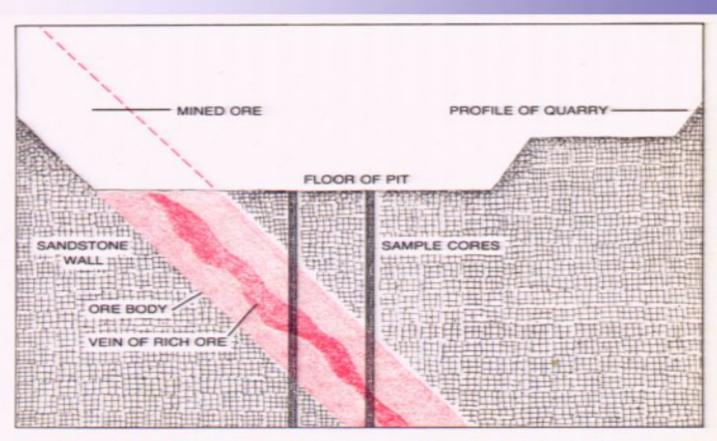






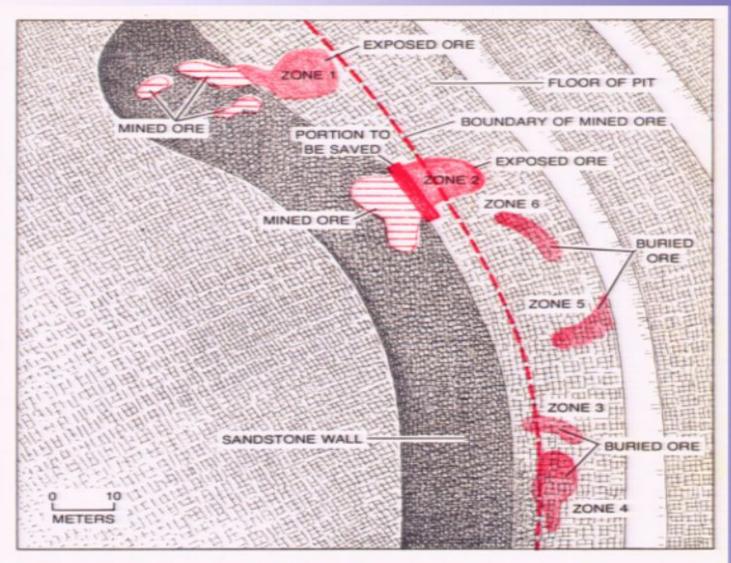


Recovery of ore samples

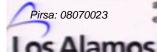


ORE BODY AT OKLO, shown in a schematic profile of the mine, lies above a sandstone wall with an average slope of about 45 degrees. The reactor zones are within the rich vein of ore, which is roughly a meter thick. The zones generally have a lenticular cross section. The extent of the ore body and the location of the reactor zones were revealed by removing core samples.

Distribution of several reactor sites



SIX REACTOR ZONES at Oklo were found in lenses of exceptionally rich ore. Parts of zones 1 and 2 had already been mined when the reactor was discovered, and their boundaries are therefore conjectural. Zones 3, 4, 5 and 6 have not yet been exposed; they lie below the present floor of the pit and were found by drilling cores. A portion of Zone 2 has been pinned to the rock face so that it will not be destroyed when the pit is deepened. In ore from the reactor zones the average concentration of uranium is from 20 to 30 percent, more than 50 times the concentration elsewhere. On the other hand, the uranium in the reactor zones is strongly depleted in U-235.



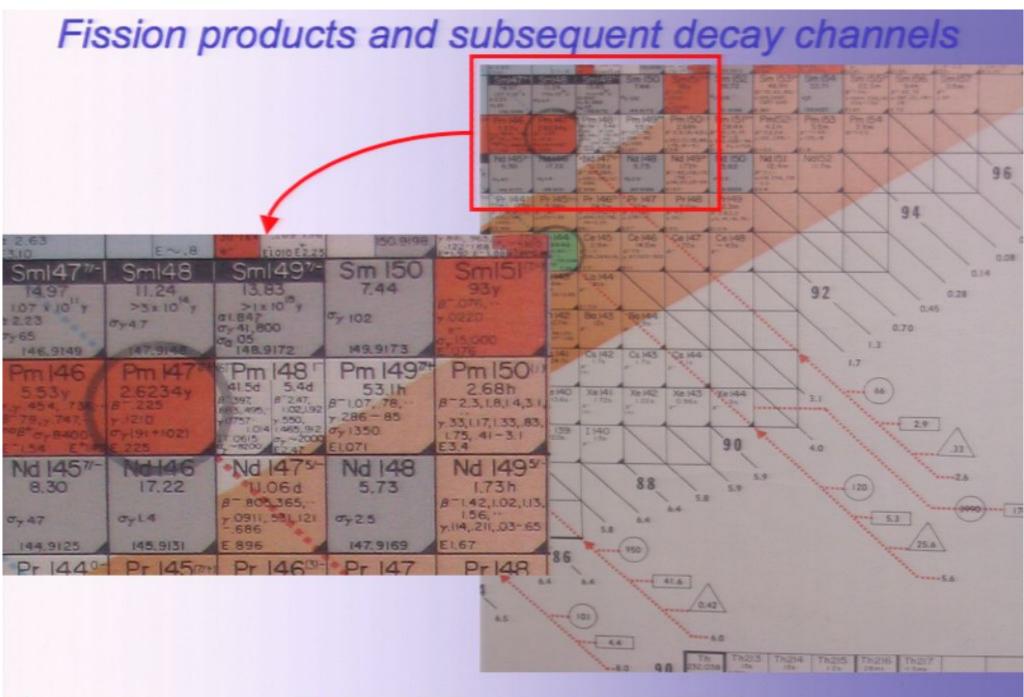
Operating characteristics of Oklo reactors

- Operating lifetime estimated as about 100,000 years: of sufficient duration for ²³⁹Pu, bred by ²³⁸U neutron absorption, to decay to ²³⁵U and contribute to the reaction
- Some samples actually show enhanced ²³⁵U isotopic abundance
- Average age of reactors: 1.8 x 10⁹ years
- Integrated neutron flux: Φ~10²¹/cm²
- Recent work indicates the reactor operated at low temperature (Meshick, Wash. U.)
 - Operation not continuous: chain reaction supported for only a few minutes
 - Long cycle of cooling between successive chain reactions









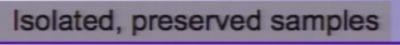


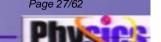


Retention of fission products at Oklo

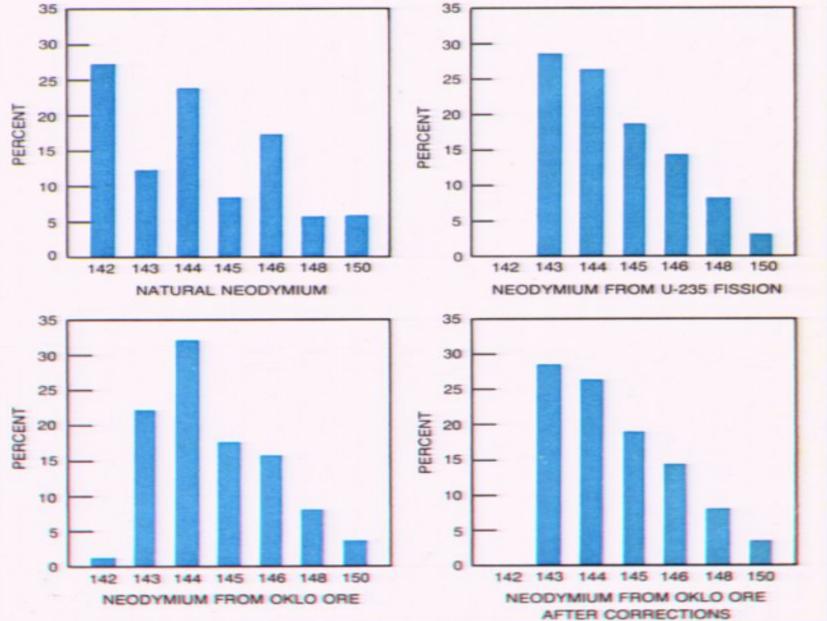
1 H 3 Li	4 Be		Retained		Partially retained Local redistribut					5 B	60	7 N	8	9 F	2 He 10 Ne		
11 Na	12 Mg		Mobilized							13 Al	14 Si	15 P	16 S	17 a	18 Ar		
19 K	20 Ca	21 Sc	Z2 Ti	23 V	24 Cr	25 Mn	26 Fe	27 Co	28 Ni	29 Ou	30 Zn	31 Ga	32 Ge	33 As	34 Se	35 Br	36 Kr
37 Rb	38 Sr	39 Y	40 Zr	41 Nb	42 Mo	43 Tc	44 Ru	45 Rh	46 Pd	47 Ag	48 Cd	49 Sb	50 Sn	51 Sb	52 Te	53 	54 Xe
55 Cs	56 Ba	57- 71	72 Hf	73 Ta	74 W	75 Re	76 0s	77 Ir	78 Pt	79 Au	80 Hg	81 TI	82 Pb	83 Bi	84 Po	85 At	86 Rn
87 Fr	88 Ra																
			57 La	58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	ස Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu
		L	89 Ac	90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md	102 No	103 Lw





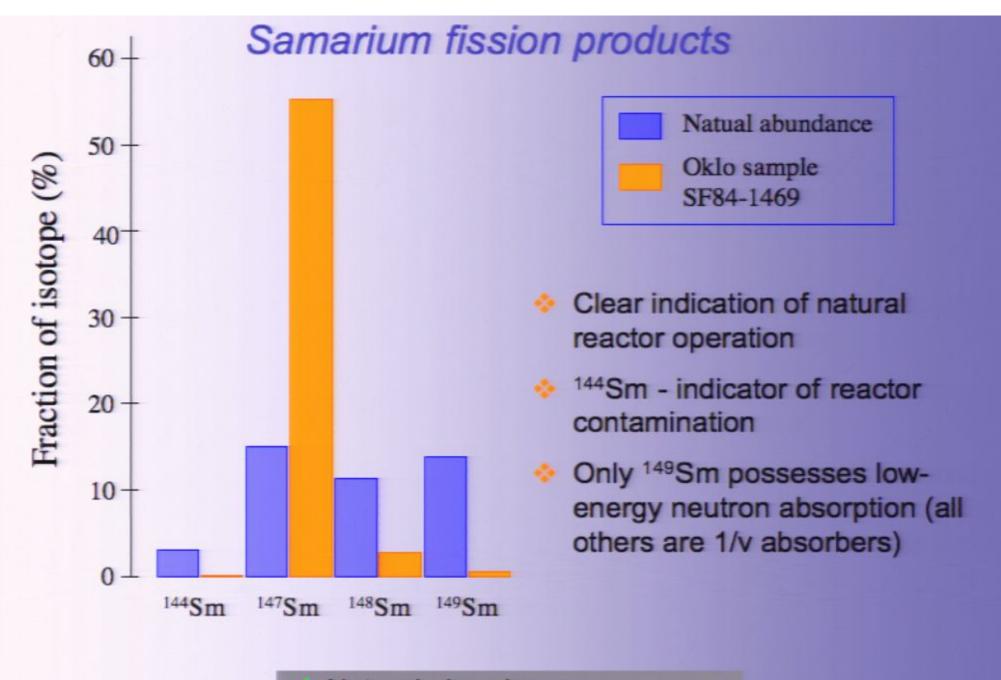


Detection of fission products from ore





Physics



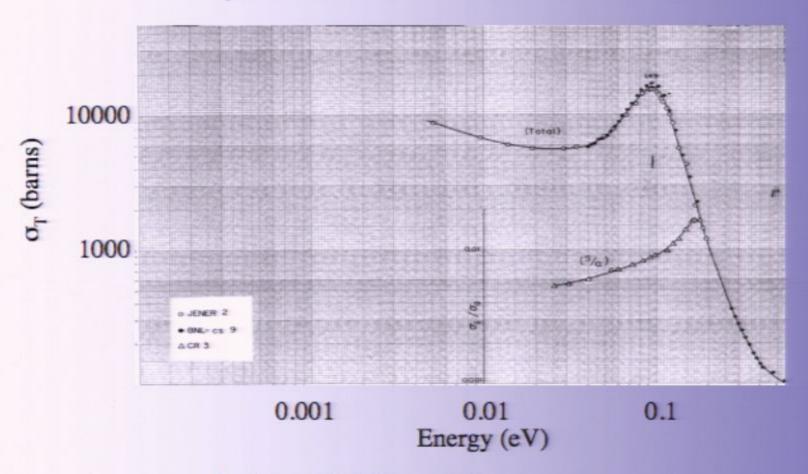


Natural abundances

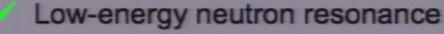
Reactor-produced abundances



Experimental 149Sm resonance



- $E_r = 97.3 \text{ meV}$
- Reproduced by several groups



Effect of neutron captures



Calculation of cross-sections from neutron spectra

Breit-Wigner approximation:

$$\sigma(E) = \frac{g\hbar^2\pi}{2mE} \frac{\Gamma_n \Gamma_{\gamma}}{\left(E - E_r\right)^2 + \Gamma_{tot}^2/4}$$

$$g, \Gamma_{n,0}, \Gamma_{\gamma} \text{ all available in tabularized form}$$

$$\Gamma_{tot} = \Gamma_n(E) + \Gamma_{\gamma}$$

$$E \text{ known from experiment}$$

$$\Gamma_n(E) = \sqrt{E}\Gamma_{n,0}$$

- tabularized form
- E, known from experimental data

$$\langle \sigma \rangle = \int \sigma(E) \Phi_n(E,T) dE \implies \langle \sigma \rangle = \frac{\int \sigma(E) \Phi_n(E,T) dE}{v_0 \int \rho_n(E,T) dE}$$

For 1/v absorber (eg. ²³⁵U):

$$\langle \sigma \rangle = \sigma(E_0)$$
 Φ

$$\Phi_n(E,T) = v_0 \int \rho_n(E,T) dE$$



Calculation of cross-sections from Oklo samples

$$\frac{dN_{147}}{dt} \frac{1}{\Phi_n} = -\langle \sigma_{147} \rangle N_{147} + \langle \sigma_{5,f} \rangle Y_5^{147} N_5^0 e^{-\langle \sigma_a \rangle \Phi_n t}$$

$$dN_{147} = -\langle \sigma_{147} \rangle N_{147} + \langle \sigma_{5,f} \rangle Y_5^{147} N_5^0 e^{-\langle \sigma_a \rangle \Phi_n t}$$

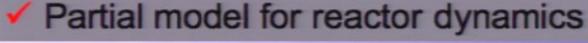
$$\frac{dN_{148}}{dt}\frac{1}{\Phi_n} = \langle \sigma_{147} \rangle N_{147}$$

$$\frac{dN_{149}}{dt} \frac{1}{\Phi_n} = -\langle \sigma_{149} \rangle N_{149} + \langle \sigma_{5,f} \rangle Y_5^{149} N_5^0 e^{-\langle \sigma_a \rangle \Phi_n t}$$

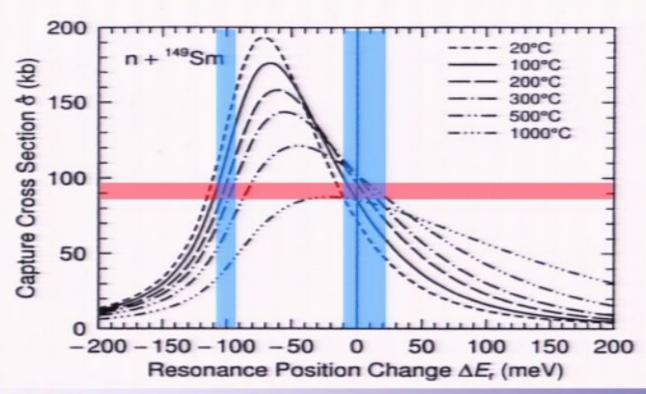
	Sample						
	SF84-1469	SF84-1480	SF84-1485	SF84-1492	SD.37		
$\hat{b}t_1$ (1/kb)	0.525	0.798	0.622	0.564	0.780		
$V_{144}(t_1)$ (%)	0.1052	0.2401	0.2073	0.1619	0.06909		
$V_{147}(t_1)$ (%)	55.34	53.23	54.03	54.81	52.74		
$V_{148}(t_1)$ (%)	2.796	3.468	3.079	2.890	4.694		
$N_{149}(t_1)$ (%)	0.5544	0.2821	0.4466	0.4296	0.3088		
$N_{235}(t_1)/N_{238}(t_1)$	0.03181	0.02665	0.02971	0.03047	0.02435		
$\hat{\tau}_{149}$ (kb)	85.6	96.5	83.8	99.0	89.5		







Results of Fujii, et al.



- Uses Maxwell-Boltzman distribution for $\Rightarrow \rho_n(E,T)$ (assumes thermal equilibrium)
- \Leftrightarrow Experimentally determined $\Rightarrow \langle \sigma_{149} \rangle$ (from these data)
- Two solutions: $\Delta E = -(97 \pm 8) \times 10^{-3} eV$ $\Delta E = +(4 \pm 16) \times 10^{-3} eV$



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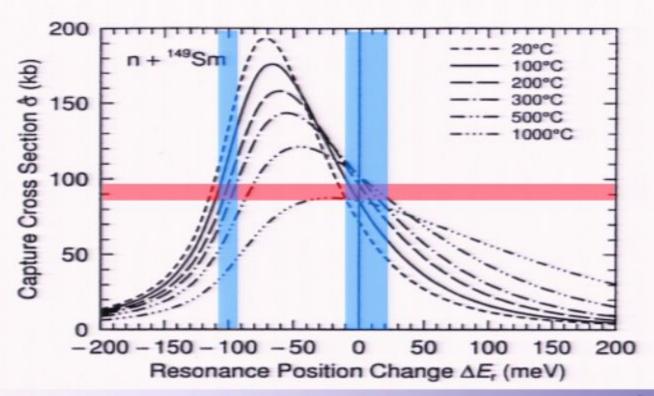
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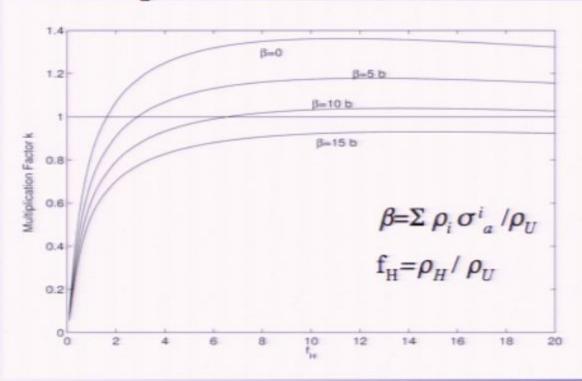
$$\Delta E = +(4 \pm 16) \times 10^{-3} eV$$





Homogeneous reactor with H₂O moderator

Multiplication factor for 3.7% ²³⁵U



$$k = vp \frac{x\rho_U \sigma_f^{235}}{x\rho_U \sigma_a^{235} + (1-x)\rho_U \sigma_a^{238} + \rho_H \sigma_a^H + \sum \rho_i \sigma_a^i}$$

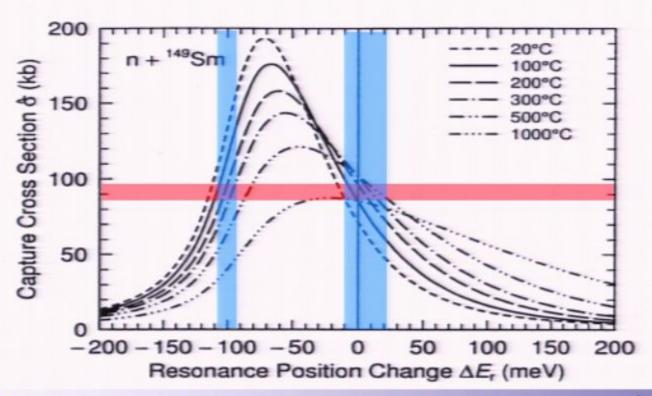
v = 2.47 Neutrons per fission

$$p = e^{-0.7(\rho_U/\rho_H)^{0.58}}$$
 Resonance escape probability





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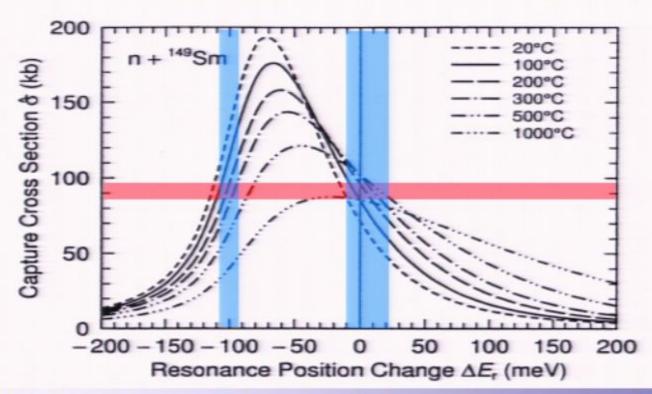
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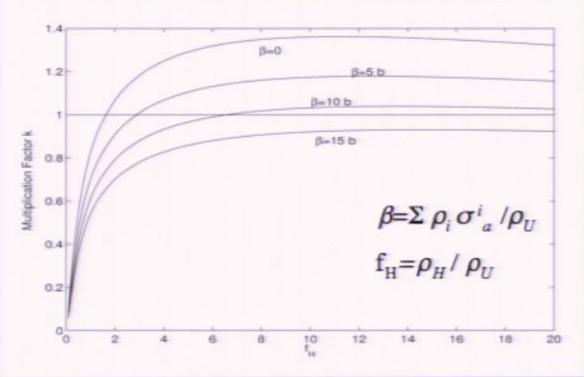
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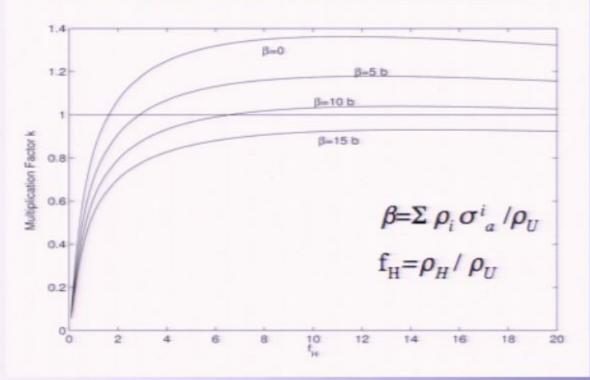
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Homogeneous reactor with H₂O moderator

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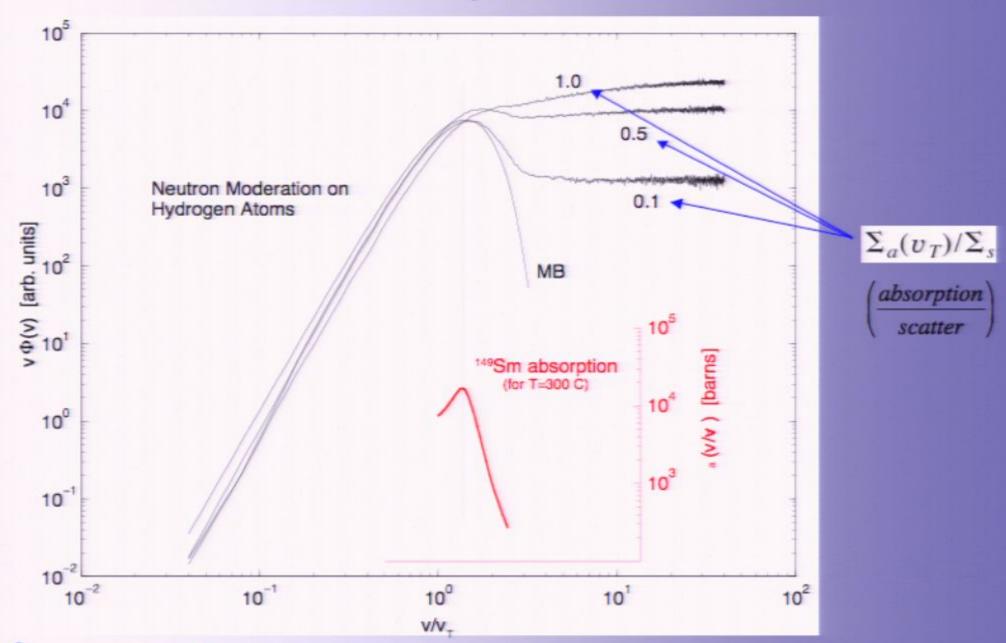
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 Resonance escape probability





Realistic neutron spectra with absorbers



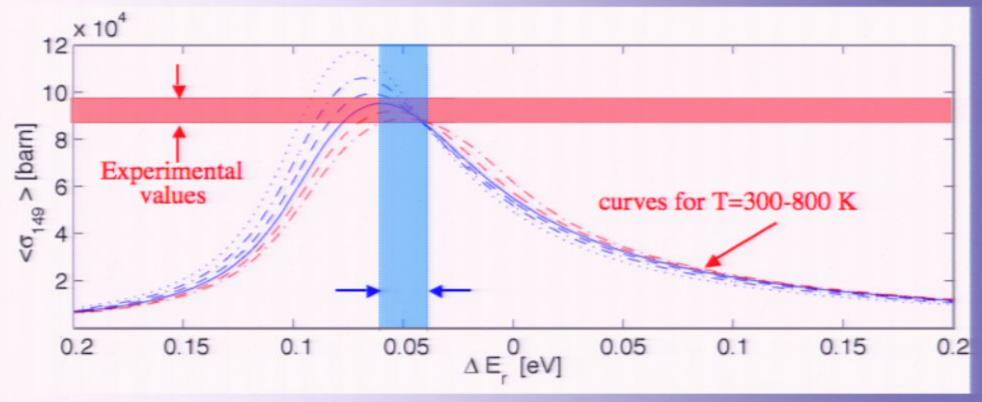
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Los Alamos

Our first results

Calculated $\langle \sigma_{149} \rangle$ as a function of resonance energy



$$\Delta E = -\left(45 \frac{+7}{-15}\right) \times 10^{-3} eV$$

$$\frac{\Delta E}{-1.1 MeV} = \frac{\Delta \alpha}{\alpha} \qquad \frac{\Delta \alpha}{\alpha} = + \begin{pmatrix} 45 + 17 \\ 45 - 7 \end{pmatrix} \times 10^{-9}$$





More complete calculation of total cross section

Including effects from ²³⁹Pu

$$\frac{dN_{147}}{dt} \frac{1}{\Phi_n} = -\langle \sigma_{147} \rangle N_{147} + \langle \sigma_{5,f} \rangle Y_5^{147} N_5 + \langle \sigma_{9,f} \rangle Y_9^{147} N_9$$

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More complete approach to uranium burning:

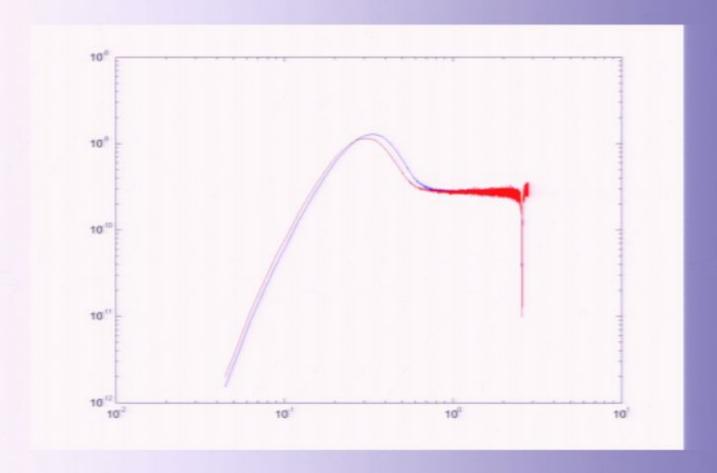
$$\frac{dN_8}{dt} \frac{1}{\Phi_n} = -\langle \sigma_8 \rangle N_8 - \Gamma \qquad \qquad \Gamma = (1 - p) \Big(v_9 \langle \sigma_{f,9} \rangle N_9 + v_5 \langle \sigma_{f,5} \rangle N_5 \Big)$$

$$\frac{dN_5}{dt} \frac{1}{\Phi_n} = -\langle \sigma_5 \rangle N_5 + \frac{\lambda_9}{\Phi_n} N_9 \qquad \qquad \frac{dN_9}{dt} \frac{1}{\Phi_n} = \langle \sigma_8 \rangle N_8 - \langle \sigma_9 \rangle N_9 - \frac{\lambda_9}{\Phi_n} N_9$$

Physics

Pirsa: 08070023

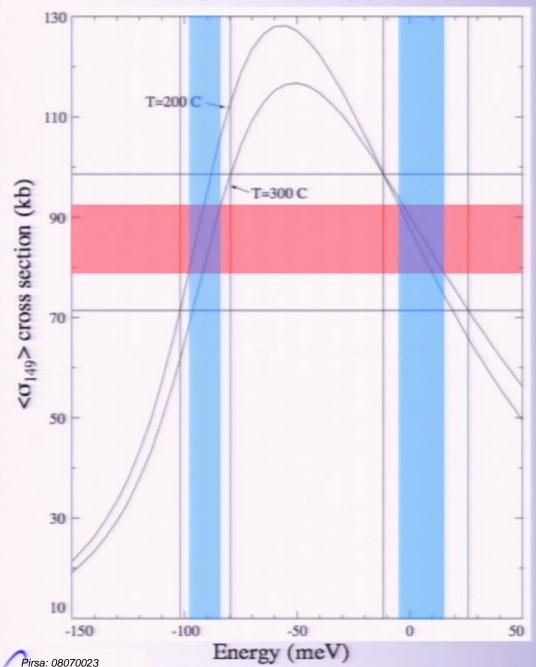
MCNP calculation of neutron spectra



- Fewer "slow" neutrons
- More pronounced structure around 100 meV



MCNP calculation of neutron spectra -



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Still two solutions:

$$\Delta E = + \begin{pmatrix} 4 + 12 \\ -9 \end{pmatrix} \times 10^{-3} eV$$

$$\Delta E = -\left(91^{+8}_{-6}\right) \times 10^{-3} eV$$

- Some issues:
 - Finite temperature of absobers
 - Chemical potentials (ie. Protons in H₂0 are treated as free)
 - Inhomogeneous reactor
 - Isotope-dependent chemistry (aging of samples over 2 billion years)

Conclusion

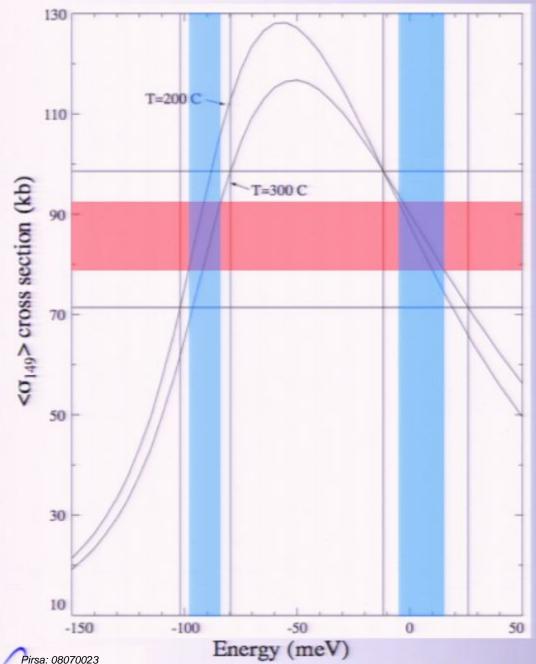
- Ideally, Oklo Phenomenon can be a sensitive test for time variation of fundamental constants
- Result is model-dependent
- Finite-size restriction produced a significant change to the values for ΔE_r
 - For example: chain reactions were found to be limited to a few 10cm within the ~1m x 3m reactor sites
- Estimates of un-included effects suggest that the result will not change by more than 10%
- Data from 155Gd is poor due to contamination with natural Gd, but favor the lower energy bound near ΔE_{r} (some bias in this determination)
- Need a better model to set bounds on fundamental parameters:

$$\frac{d\ln(E_r)}{dt} = c_{\alpha} \frac{d\ln(\alpha)}{dt} + c_{\Lambda} \frac{d\ln(m_q/\Lambda_{QCD})}{dt}$$





MCNP calculation of neutron spectra -



los Alamos

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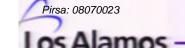
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_ Phue

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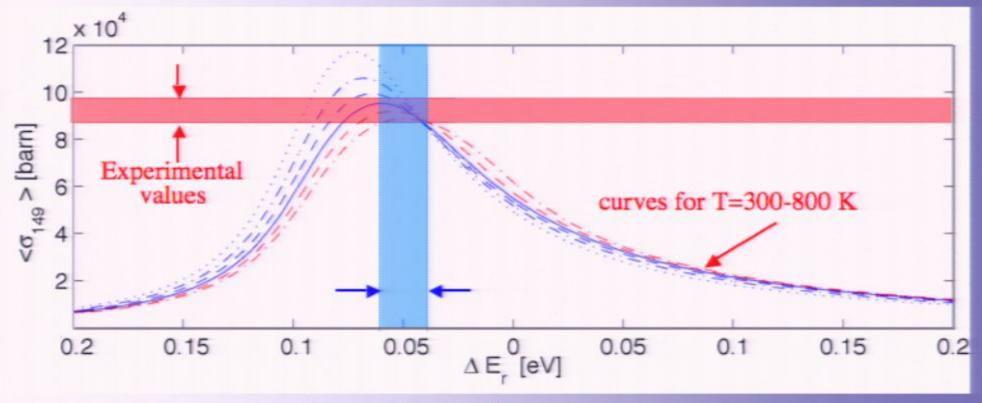
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Physical Physical

History of searches for time variation of α with Oklo

Basic idea due to A.I. Shlyakhter, Nature 264, 340 (1976) $\left|\frac{\dot{\alpha}}{\alpha}\right| < 1 \times 10^{-17}/yr$

- More complete analysis by Damour and Dyson, Nucl. Phys. B 480, 37 (1996)
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- Y. Fujii et al. obtained better samples, six times better precision, Nucl. Phys. B 573, 377 (2000)

 $\frac{\dot{\alpha}}{\alpha} = -(2\pm 8) \times 10^{-18} / yr$

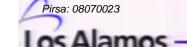
Lamoreaux and Torgerson use improved moderated neutron spectrum, Phys. Rev. D 69, 121701(R) (2004)

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Gould, Sharapov, Lamoreaux calculate neutron spectrum with sophisticated MCNP code, Phys. Rev. C 74, 024607 (2006)

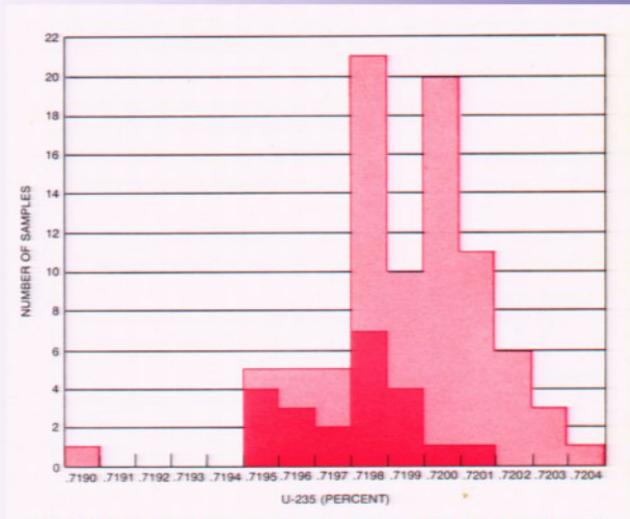
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Worldwide 235U fraction



ABUNDANCE OF U-235 in 88 samples of uranium ore shows a clearly bimodal distribution. The peak at .7200 percent represents ores mined mainly in Canada, Europe and Australia. The peak at .7198 percent is produced by ores taken from the Colorado Plateau region of the U.S. One group of these ores, mined in New Mexico, is shown separately (dark color); it plainly suggests that uranium from the Colorado Plateau has a U-235 content smaller than the world average. One possible explanation of this small discrepancy is that the isotopes were separated by some chemical process in the ore-bearing rocks. Another explanation is that a natural reactor once operated in the region and that its depleted ore has been dispersed throughout the plateau.



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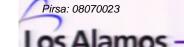
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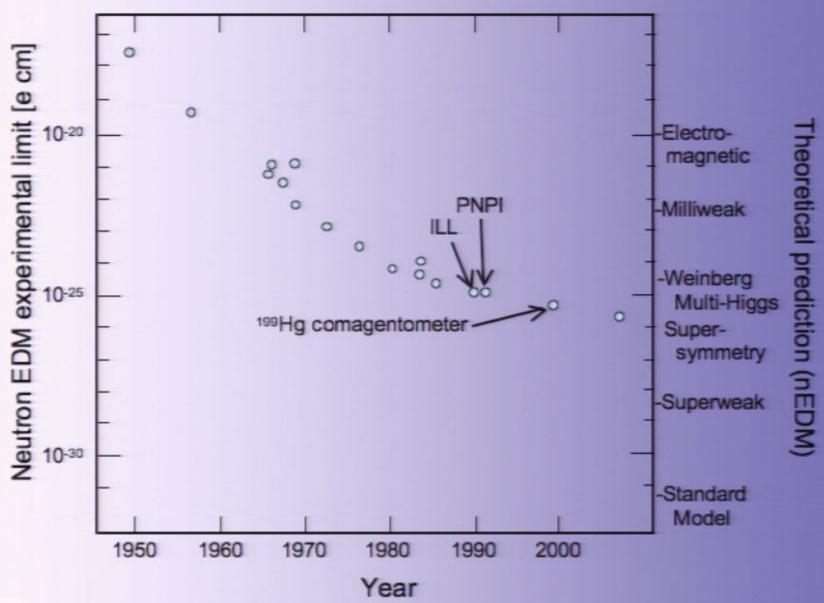
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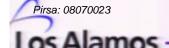
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Evolution of searches for EDMs





- Physics

