Transactinium elements in the evolving universe

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An exponential model of continuous galactic synthesis is used to analyze new data on the abundance in meteorites, the Earth, and the moon of several transactinium nuclei, particularly plutonium-244. It is shown that nucleosynthesis in our galaxy occurred over a period of six billion years up to the formation of the solar system. The possibility of a change in nuclear stability in the past as the result of change in the universal constants is also discussed. It is shown that this possibility is greatly limited but that a direct check of the constancy of the constants could be obtained by comparison of the radii of pleochroic rings in old micas with contemporary α-particle ranges.

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CONTENTS

INTRODUCTION

In recent years interesting new facts have appeared in cosmology which should substantially influence the current ideas of the evolution of the observed universe as did the discovery of quasars and the three-degree microwave radiation in their time. These facts are related, however, not to optical or radio astronomy observations but are due to investigation of the abundance in the solar system of certain radioactive nuclei and particularly of the transactinium nuclides such as ^{232}Th, ^{235}U, ^{238}U, ^{237}Np, ^{244}Pu, and ^{247}Cm.

The existence in the universe of appreciable quantities of radioactive nuclei, particularly of heavy nuclei, is one of the direct proofs that the universe has changed and is changing continuously in its composition. This was first pointed out in 1904 by Rutherford,[1] who utilized analysis of the radioactive decay of uranium and the ratio of concentrations of helium and uranium in rocks to determine the age of a piece of uraninite, i.e., the period of time which had passed from the moment of solidification of the uraninite, when the helium stopped leaving it. As has been noted recently by Clayton[2] this analysis can be considered historically as the first fact regarding the evolution of the universe.

The accumulation of observational data (Hubble's law, 1929) and the application of the equations of general relativity to the entire universe (the homogeneous isotropic models of Friedmann, 1922 and 1924, LeMaitre, 1927, and others) have led to the conclusion that the universe is a nonstationary expanding system. Here, according to Friedmann's theory, the evolution of the universe with time depends substantially on the value of the current average density of matter for a given rate of expansion. For a density $\rho_{curr} < \rho_{cr}$ the homogeneous universe is infinite and will expand without limit (the open model); for a density $\rho_{curr} > \rho_{cr}$ the universe is closed, its volume is finite, and expansion should be replaced by compression (the closed model). Regardless of the nature of the expansion, it is clear that it cannot have continued indefinitely in the past and at some moment $t = 0$ the density of matter must have been infinite ($\rho \to \infty$).[1)]

Gamow in 1946 proposed that the universe in the early stages near the singularity was sufficiently hot that thermonuclear reactions of helium synthesis could occur in it.

Three well known observational facts exist which favor the hot model of the universe (the big-bang model). The first is the discovery of the black-body radiation remaining from the hot universe. The second is the fact that the ages of the oldest stars and the ages of the spherical clusters are $\sim(11-13)\times 10^9$—in agreement with the age of the universe determined in relativistic cosmological models as the time which has passed from the beginning of the expansion (the big bang): $t_p = (1-2)\times 10^{10}$ years. The third fact is that the age of the elements of our galaxy determined from radioactive transactinium nuclides is, as we will see below, of the same magnitude, $(11 \pm 2)\times 10^9$ years.

ORIGIN OF THE ELEMENTS

The hot model satisfactorily explains the synthesis of the simplest elements deuterium and helium in the early stages of the evolution of the universe. The synthesis of heavy elements occurs in the interiors of stars and in explosive processes—in the flashes of novae and supernovae. This mechanism of nucleosynthesis, which includes nuclear reactions and neutron-capture proces-

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[1)] In recent years it has become clear that the singularity also is not removed in extremely general inhomogeneous and anisotropic cosmological models [46].

ses, permits description of the formation of all chemical elements from helium to the transuranium elements.

Synthesis in the hot model. In the hot model[3] in the early stage of evolution,[2] $\tau \sim 10^{-6}$ sec ($\Omega = \rho_{cr}/\rho \approx 0.05$) from the beginning of the expansion, only nucleons remain among the baryons, and as the result of the strong interaction they are transformed from one to the other:

$$\left.\begin{aligned} p+\pi^- &\rightleftarrows n+\gamma, \\ n+\pi^+ &\rightleftarrows p+\gamma, \\ T &> 10^{12}\ {}^\circ\mathrm{K}. \end{aligned}\right\} \tag{1}$$

At a temperature below 10^{11} °K the quantity of mesons decreases sharply and the mutual transformation of neutrons and protons via mesons is cut off. However, as was shown by Hayashi[4] the mutual transformation of neutrons and protons is now maintained through weak interactions such as

$$\left.\begin{aligned} n+e^+ &\rightleftarrows p+\bar{\nu}_e, \\ n+\nu_e &\rightleftarrows p+e^-, \\ n &\rightleftarrows p+e^-+\bar{\nu}_e. \end{aligned}\right\} \tag{2}$$

In this case complete equilibrium exists between all forms of matter and radiation at a temperature above 10^{10} °K ($\tau < 1$ sec). For temperatures below 10^{10} °K the weak interaction is not able to preserve the thermodynamic equilibrium between neutrons and protons, since the concentration of electron-positron pairs begins to drop sharply. The ratio p/n will be frozen until neutron decay becomes important. This frozen value for $\tau \sim 10$ sec is p/n = 4.8 (17% neutrons). Formation of a deuteron from a neutron and a proton, $n + p \rightarrow D + \gamma$, is nevertheless impossible for temperatures $T > 10^9$ °K, since there are many photons capable of rapidly destroying the deuterons. At $\tau \approx 100$ sec ($T \approx 10^9$ °K) formation of deuterons and eventually helium begins:

$$\left.\begin{aligned} &\mathrm{D}+\mathrm{D}\rightarrow\begin{cases} {}^3\mathrm{He}+n, \\ {}^3\mathrm{H}+p, \end{cases} \\ &{}^3\mathrm{He}+n\rightarrow{}^3\mathrm{H}+p, \quad \mathrm{D}+p\rightarrow{}^3\mathrm{He}+\gamma, \\ &{}^3\mathrm{H}+\mathrm{D}\rightarrow{}^4\mathrm{He}+n, \quad {}^3\mathrm{H}+p\rightarrow{}^4\mathrm{He}+\gamma. \end{aligned}\right\} \tag{3}$$

At this stage the neutrons have the frozen concentration ~17% (Fig. 1). If all neutrons are converted to helium, the final concentration of helium-4 will be ≈30% by mass, in agreement with the observations. Thus, the hot model satisfactorily describes the synthesis of helium-4 in the universe. The problem of deuterium and helium-3 is being debated, but as yet there is no clear opinion.[47] However, we can now say with complete reliability that this model does not explain the existence of elements heavier than helium (except perhaps for ^{7}Li, whose content in the solar system is only 10^{-6}%).

The absence of stable nuclei with atomic weight 5 and 8 in nature inhibits, in terms of the hot model, the formation of elements heavier than helium, since the low density of matter does not allow this gap to be crossed by means of multiple (perhaps ternary) collisions. Therefore we must turn to noncosmological processes for formation of elements heavier than helium. Since the atoms of these elements comprise only 0.1% of all atoms of the universe, we can assume that their synthesis occurred much later in the interior of stars formed from the primary hydrogen and helium.

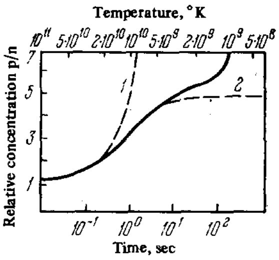

FIG. 1. Variation with time of the proton-neutron ratio p/n. The solid curve reflects the real behavior of this ratio with time; dashed curve 1 corresponds to thermodynamic equilibrium between neutrons and protons, $p/n = \exp(\Delta mc^2/kT)$; curve 2 reflects the behavior of p/n when the free decay of neutrons can be neglected.

As hydrogen, helium, carbon, and silicon are burned up in the dense nuclei of giant stars, synthesis occurs of all heavier nuclei up to iron. A detailed review of thermonuclear synthesis in stars has been published by Clayton.[5]

Nucleosynthesis in stars and the problem of rhenium-187. We shall discuss here only the last stage of this synthesis, which leads to formation of heavy and transactinium elements. In this stage, synthesis of the elements occurs by means of slow capture of neutrons in which β-decay processes can occur (the s process), and also by the rapid capture of neutrons in which β-active nuclei are not able to decay (the r process).

If the nuclei of atoms of heavy elements are produced not as the result of random reactions not associated with each other, but arise in a single nucleosynthesis process,[3] then the knowledge of their relative terrestrial abundances and rates of decay will permit determination of the period of time which has passed from the moment of termination of the nucleosynthesis. On the other hand, the models of the s and r processes permit description of the time dependence of the synthesis of the elements. The duration of nucleosynthesis in the galaxy Δ also is of considerable interest, since it can be directly determined (without additional assumptions) from the long-lived isotopes.

In this respect, of the elements formed in the s process the most remarkable is osmium-187, 60% of which on the Earth must be considered as originating from the β decay of rhenium-187 (half-life $T_{1/2} = 4.3 \times 10^{10}$ years,[6] decay energy 2.6 keV). Measurements of the cross sections for neutron capture have now confirmed the correctness of the theory of the s process, on the basis of which this conclusion was drawn.

According to the theory of the s process the contents (yields) of nuclei in the chain of the s process for small regions of variation of A are inversely proportional to the average cross sections for capture of thermal neutrons by the given nuclei, i.e., we have the quantity $N_S(A)\,\bar{\sigma}(A) = \mathrm{const}$. In particular, $^{187}\mathrm{Os}_{[S]}\,\bar{\sigma}(^{187}\mathrm{Os}) = {}^{186}\mathrm{Os}\,\bar{\sigma}(^{186}\mathrm{Os})$, and the amount of radiogenic osmium-187 produced from rhenium-187 is

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[2] The purpose of this article is to analyze the origin of heavy elements. Cosmological mechanisms of formation of the elements have been discussed briefly, since they explain the synthesis only of the light elements.

[3] It is possible to make very different assumptions regarding the astrophysical circumstances in which this single nucleosynthesis process occurred. This could be the flash of any one supernova or a number of successive flashes of supernovae up to the formation of the solar system. According to current ideas the explosion of a supernova leads to disruption of the shell of the star, so that a significant part of the material synthesized in the explosion (when an intense neutron flux is produced) is ejected into interstellar space. The matter thrown out from stars in such flashes is mixed with the interstellar gas and, having dissolved in it, serves as further material for formation of younger stars and systems (for example, the solar system).

$$^{187}\mathrm{Os}_{[\mathrm{rg}]} = {}^{187}\mathrm{Os} - {}^{187}\mathrm{Os}_{[s]} = {}^{187}\mathrm{Os} - \left[\frac{\bar{\sigma}(^{186}\mathrm{Os})}{\bar{\sigma}(^{187}\mathrm{Os})}\right]^{186}\mathrm{Os}, \qquad (4)$$

where $^{187}\mathrm{Os}_{[rg]}$ is the number of atoms of radiogenic Os^{187}; $^{187}\mathrm{Os}_{[S]}$ is the quantity of $^{187}\mathrm{Os}$ atoms produced in the s process; $\bar{\sigma}(A)$ is the average cross section for neutron capture for nuclei of mass A; $^{186}\mathrm{Os}$ is stable and does not have a radiogenic origin (it is formed only in the s process).

From Eq. (4), if we take into account the measurement[7] $\bar{\sigma}(^{186}\mathrm{Os})/\bar{\sigma}\,^{187}\mathrm{Os} = 0.4 \pm 0.1$ and $^{186}\mathrm{Os}/^{187}\mathrm{Os} = 1$, we obtain

$$\frac{^{187}\mathrm{Os}_{[\mathrm{rg}]}}{^{187}\mathrm{Os}} = 0{,}6\,(60\%), \qquad (5)$$

$$\frac{^{187}\mathrm{Os}[\mathrm{rg}]}{^{187}\mathrm{Re}} = \frac{(^{187}\mathrm{Os}/\mathrm{Os}) - [\bar{\sigma}(^{186}\mathrm{Os})/\bar{\sigma}(^{187}\mathrm{Os})]\,(^{186}\mathrm{Os}/\mathrm{Os})}{^{187}\mathrm{Re}/\mathrm{Re}}\,\frac{\mathrm{Os}}{\mathrm{Re}} = 0.12 \pm 0.03. \qquad (6)$$

Since this quantity corresponds to the decay of $^{187}\mathrm{Re}$ for a period of approximately a quarter of its half-life ($\approx 10^{10}$ years), the radiogenic nature of $^{187}\mathrm{Os}$ seems astonishing in the light of current ideas of the age of the solar system. Is it possible that the $^{187}\mathrm{Os}$ was produced long before the formation of the sun? As we will see in what follows, this is inconsistent with contemporary models of nucleosynthesis.

Clayton recently made the interesting suggestion[8] that the rate of decay of $^{187}\mathrm{Re}$ can increase under certain astrophysical circumstances, particularly for high stellar temperatures, $10^5 < T < 10^8$ °K. Thermal population of the excited states of $^{187}\mathrm{Re}$ (134 keV and above) and their subsequent decay to $^{187}\mathrm{Os}$ cannot, however, lead to such an acceleration, since even at $T = 10^8$ °K this effect is very weak: the transition $7^{\pm}/2 \rightarrow 1^{-}/2$ corresponds to $\log ft \approx 9$. The β decay induced by photons is also too weak at these temperatures.

The increase in the rate of decay of $^{187}\mathrm{Re}$ can be explained only as the result of an additional mechanism of β decay to bound states of the partially ionized atom. In β decay to a bound atomic state, the electron originating in the nucleus fills a vacant position in one of the atomic shells. Brodzinski and Conway[9] showed that about a third of the decays of terrestrial $^{187}\mathrm{Re}$ occur to a vacant bound state in the $^{187}\mathrm{Os}$ daughter atom.

In the interiors of stars, where the atoms are highly ionized, such β decay to bound atomic states will be accelerated as the result of the increase in the number of vacant states of electrons in the atom because of ionization, and also as a result of the fact that decay to vacancies of the inner shells of an ionized atom will occur more rapidly than decay to vacancies of the outer shells of a neutral atom.

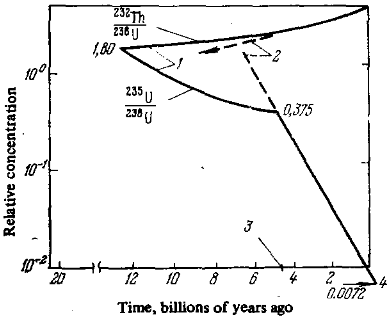

FIG. 2. Variation with time of the relative concentrations $^{232}\mathrm{Th}/^{238}\mathrm{U}$ and $^{235}\mathrm{U}/^{238}\mathrm{U}$. 1–Continuous synthesis, 2–sudden synthesis, 3–formation of solar system, 4–relative concentration $^{235}\mathrm{U}/^{238}\mathrm{U}$ at the present time.

In the sun at a temperature of 10^6 °K rhenium will contain bound electrons in the M shell, but all subsequent outer shells beginning with N will be ionized. This atom of rhenium has 28 electrons and 48 vacant states for the electron arising in β decay. If we assume that the rate of β decay of $^{187}\mathrm{Re}$ to bound atomic states is increased in the sun by at least 48 times (the number of new final states), in this case the half-life of $^{187}\mathrm{Re}$ can decrease from 43×10^9 years (the terrestrial value) to 2.6×10^9 years, which is less than the age of the sun.4)

Measurement of the isotopic composition of $^{187}\mathrm{Re}$ and $^{187}\mathrm{Os}$ in the solar wind could serve as a verification of Clayton's idea. If the idea proposed above for the mechanism of decay of rhenium-187 is valid, the solar wind should be strongly depleted in rhenium and enriched in osmium.

If this is not the case, then the great age of $^{187}\mathrm{Re}$ leads to a contradiction with recent cosmochronological data on the transactinium elements.

"Time, gentlemen, please!".[11]

NUCLEAR COSMOCHRONOLOGY. THE ROLE OF TRANSACTINIUM ELEMENTS

In this chapter we discuss the time scale of nuclear synthesis and the formation of the solar system, based on data on concentrations in the Earth and in meteorites and yields in nuclear synthesis of radioactive transactinium nuclides, and also of iodine-129.

Up to 1971 only three long-lived natural transactinium nuclides were known on the Earth,5) U^{235}, U^{238}, and Th^{232}. To determine the age of these three nuclides it is necessary to know their relative abundance in the solar system at the present time and also the rate of production in the r process.

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4)The theory of β decay to bound atomic states has been discussed by Bahcall [10] in application to allowed transitions. If we consider the the quantity Γ_b/Γ_c–the ratio of probabilities of β decay to bound states (Γ_b) and to continuum states (Γ_c), then Γ_b/Γ_c will be determined mainly by the phase spaces for decays respectively to bound states and to the continuum. The phase space of β decay to the continuum is represented by a certain function $f(Z, W_0)$, where W_0 is the decay energy and Z is the nuclear charge. For decays to bound states the analogous phase space is the product of the square of the neutrino momentum and the square of the modulus of the electron wave function calculated at the surface of the nucleus. Hence the *relative* probability of β decay to bound atomic states is

$$\frac{\Gamma_b}{\Gamma_c} \sim q^2 \frac{|\psi_e(R)|^2}{f(Z, W_0)}.$$

Under terrestrial conditions atoms are neutral, and only one vacant state exists for an electron arising in β decay; therefore the relative probability of β decay to bound states is small (for example, for the decay of tritium we have $^3\mathrm{H} \xrightarrow{\beta^-} {}^3\mathrm{He}$ $\Gamma_b/\Gamma_c \approx 6.9 \cdot 10^{-3}$), and the half-life is determined by the probability of β decay to the continuum: $T_{1/2} \sim 1/(\Gamma_b + \Gamma_c) \approx 1/\Gamma_c$. If about a third of the decays of terrestrial $^{187}\mathrm{Re}$ occurs to one vacant bound state in the daughter atom $^{187}\mathrm{Os}$, this means that on the Earth $\Gamma_b/\Gamma_c = 0.5$. If on the sun $\Gamma_b' = 48\Gamma_b$, then $\Gamma_b'/\Gamma_c = 24$. In this unique case the half-life will be determined mainly by decay to bound atomic states (!):

$$\frac{T_{1/2*}}{T_{1/2\odot}} = \frac{(\Gamma_b'/\Gamma_c)+1}{(\Gamma_b/\Gamma_c)+1} \approx 17.$$

5)All three nuclides undergo α decay: $\xrightarrow{T_{1/2} = 7{,}1 \cdot 10^8\ y} {}^{231}\mathrm{Th} \rightarrow \ldots,$ $^{238}\mathrm{U} \xrightarrow{T_{1/2} = 4{,}5 \cdot 10^9\ y} {}^{234}\mathrm{Th} \rightarrow \ldots,$ $^{232}\mathrm{Th} \xrightarrow{T_{1/2} = 1{,}4 \cdot 10^{10}\ y} {}^{228}\mathrm{Ra} \rightarrow \ldots$

In nuclear cosmochronology we ordinarily utilize the change with time of the relative concentrations ^{232}Th/^{238}U, ^{235}U/^{238}U, and also ^{244}Pu/^{238}U, ^{237}Np/^{238}U, and ^{247}Cm/^{238}U.

In Fig. 2 we have shown an example of this change for ^{235}U/^{238}U and ^{232}Th/^{238}U.

In the period of galactic nucleosynthesis before the formation of the solar system the ratio of concentrations changes as the result of production of elements in the r process, and after the formation of the solar system, free decay occurs. All of the transactinium elements enumerated above are formed in the r process (Fig. 3).

In order that nuclei with a number of protons Z + 1 be formed in the r process from nuclei with a number of protons Z, the following conditions must be satisfied:

$$\left.\begin{array}{l} 1)\ \lambda_\beta(Z, N) > \lambda_{sf}(Z, N), \\ 2)\ \lambda_\beta(Z, N) > \lambda_{nf}(Z, N), \\ 3)\ \lambda_{n\gamma}(Z+1, N-1) > \lambda_{nf}(Z+1, N-1), \\ 4)\ \lambda_{n\gamma}(Z+1, N-1) > \lambda_\beta(Z+1, N-1), \end{array}\right\} \tag{7}$$

where λ_β, $\lambda_{n\gamma}$, λ_{sf}, and λ_{nf} are the rate (in sec^{-1}) respectively of β decay, neutron capture, spontaneous fission, and neutron fission.

The yields of the transactinium elements important for nuclear cosmochronology have been calculated[12] taking into account the α-decay branches and the fraction of spontaneous fission of the parent nuclei. For example, the yield of thorium-232 is determined by the combined contribution of nuclides with

$A = 232 + 4n$	$(n = 0.\ 1.\ 2.\ 3...)$
232	0.0130
236	0.0114
240	0.0167
244	0.0145
248·0.971	0.0122
252·0,892	0.0129
256·0.107	0.0005 (in rel. un.)
^{232}Th	– 0.0812

According to this scheme the following ratios of the yields of the different transactinium elements are obtained in the r process:

$$^{232}\mathrm{Th}/^{238}\mathrm{U} = 1.78,\ ^{235}\mathrm{U}/^{238}\mathrm{U} = 1.80\ ^{6)},\ ^{244}\mathrm{Pu}/^{238}\mathrm{U} = 0.88,\ ^{237}\mathrm{Np}/^{238}\mathrm{U} = 1.70. \tag{8}$$

These are the relative rates of production of the transactinium elements in nucleosynthesis and at the same time these are the initial concentrations at the moment of formation of the galaxy (the beginning of nucleosynthesis).

Chronological models of nucleosynthesis in their simplest form express the number N_A of nuclei with mass number A in the interstellar medium as a continuous function of time during nuclear synthesis from t = 0 to t = Δ:

$$\frac{dN_A(t)}{dt} = a_A(t) - \lambda_A N_A(t), \quad 0 \leqslant t \leqslant \Delta, \tag{9}$$

where $a_A(t)$ is the rate of production and λ_A is the rate of radioactive decay of the nucleus discussed with mass A; $N_A(t)$ is the number of nuclei at the moment of time t. Since we are concerned here mainly with the r process of nucleosynthesis, t = 0 is the beginning of this process in the galaxy, and t = Δ is the last moment of time when the r process contributes to formation of the material of the solar system (Fig. 4).

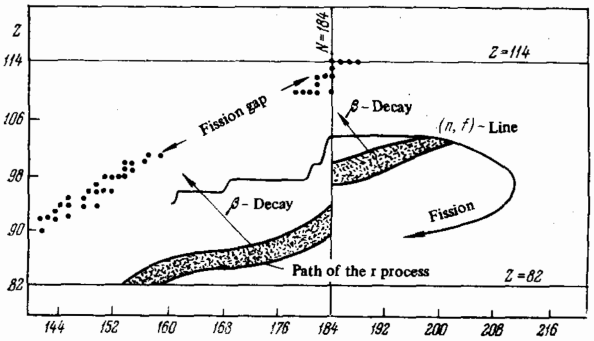

FIG. 3. Path of the r process for transactinium nuclides.

We shall designate by δ the time between the formation of the material of the solar system and the solidification of meteoritic material. The formation of meteoritic and planetary material signifies the formation of a closed system without further changes in the concentrations of the elements except for changes due to radioactive decay, for example, the decay $^{129}\mathrm{I} \rightarrow {}^{129}\mathrm{Xe}$ or the decay $^{244}\mathrm{Pu} \rightarrow {}^{232}\mathrm{Th}$. If θ_M is the age of the meteorites, then the age of the solar system θ_{SS} is related to δ as follows:[7]

$$\theta_{SS} = \theta_M + \delta. \tag{10}$$

Integration of Eq. (9) gives

$$N_A(t) = e^{-\lambda_A t} \int_0^\Delta a_A(t)\, e^{\lambda_A t}\, dt. \tag{11}$$

At the moment of solidification of the meteorites (taking into account also the free decay in the time δ) we obtain

$$N_A(\Delta + \delta) = e^{-\lambda_A(\Delta+\delta)} \int_0^\Delta a_A(t)\, e^{\lambda_A t}\, dt. \tag{12}$$

Since, as we shall show below, the age of the galaxy is

$$\theta_G = \Delta + \theta_{SS}, \tag{13}$$

the number of nuclei with mass A at the present time is

$$N_A(\Theta_G) = e^{-\lambda_A(\Delta+\theta_{SS})} \int_0^\Delta a_A(t)\, e^{\lambda_A(t)}\, dt. \tag{14}$$

The function $a_A(t)$ is determined by the type of model of nucleosynthesis. Possible types are as follows:

1) Exponential decay

$$a_A(t) = a_A(0)\, e^{-t/T^*} \tag{15}$$

(here T* is a constant independent of time which characterizes the type of nucleosynthesis),

2) uniform synthesis

$$a_A(t) = \mathrm{const}, \tag{16}$$

3) sudden synthesis

$$\begin{gathered} a_A T^* \rightarrow \mathrm{const}, \\ T^* \rightarrow 0. \end{gathered} \tag{17}$$

The exponential model is mathematically the most

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[6] Fowler [11] has pointed out that it is necessary to take into account the decrease in the yield in the r process of isotopes with odd A (for a given Z), and then the ratio ^{235}U/^{238}U is reduced to 1.42 ± 0.19.

[7] In what follows we will use the symbol M to designate the moment of solidification of metorites Δ + δ, the symbol SS to indicate the moment of formation of the solar system, and the symbol p to indicate the present time.

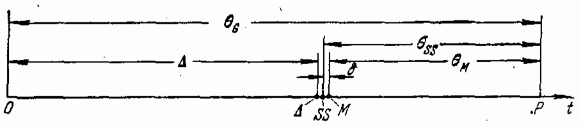

FIG. 4. Time scale of nucleosynthesis and formation of the solar system. O—Beginning of nucleosynthesis, Δ—end of nucleosynthesis—the last moment of time when the r process contributes to formation of solar system material, SS—formation of the solar system, M—formation of the meteorites and the earth, P—present time.

convenient, since it combines the three cases if we introduce a characteristic time of decrease of the rate of nucleosynthesis T*. Then T* = ∞ corresponds to uniform synthesis and T* = 0 to sudden synthesis.

Equation (12) for the exponential model leads to the relation

$$N_A(\Delta+\delta)=a_A\frac{T^*}{1-\lambda_A T^*}e^{-\lambda_A\delta}(e^{-\lambda_A\Delta}-e^{-\Delta/T^*}). \tag{18}$$

For stable nuclei ($\lambda_A = 0$) we have

$$N_A^{\text{stab}}(\Delta+\delta)=a_A T^*(1-e^{-\Delta/T^*})\equiv a_A\Delta_E, \tag{19}$$

where Δ_E is the effective duration of nucleosynthesis.

In the case of sudden explosive synthesis (T* → 0) at the moment T = 0 we have

$$N_A(\Delta+\delta)=a_A\Delta_s e^{-\lambda_A(\Delta+\delta)}, \tag{20}$$

where Δ_S is the effective duration of the explosive nuclear synthesis for t = 0.

In the case of sudden synthesis for t = Δ′ we have

$$N_A(\Delta'+\delta)=a_A\Delta_s' e^{-\lambda_A\delta}. \tag{21}$$

Finally, in the case of uniform synthesis (T* = ∞) the general Eq. (12) leads to the form

$$N_A(\Delta+\delta)=\frac{a_A}{\lambda_A}e^{-\lambda_A\delta}(1-e^{-\lambda_A\Delta}). \tag{22}$$

Let us turn now to Eq. (14). The parameters Δ and T* can be determined from this equation if we use the following values of the ratios of the abundances and rates of production of the three transactinium elements:

$$\left(\frac{^{235}\text{U}}{^{238}\text{U}}\right)_P=0.00723,\quad \left(\frac{^{232}\text{Th}}{^{238}\text{U}}\right)_P=3.8\pm0.3,\quad \frac{a_{235}}{a_{238}}=1.65\pm0.15, \tag{23}$$
$$\frac{a_{232}}{a_{238}}=1.65\pm0.15.$$

The age of the solar system $\Theta_{SS} = (4.7 \pm 0.1)\times 10^9$ years is taken independently of the other data.

The results of a calculation on the basis of these data corresponds to the "old" nuclear cosmochronological model developed by Fowler and Hoyle[12a] in 1960. Here we obtain $\Delta = 6.9\times 10^9$ years, $T^* = 11.1\times 10^9$ years for the exponential model, and $\Delta = 7.7\times 10^9$ years for the uniform model.

In recent years in nuclear cosmochronology there has not only been an improvement in the calculations of the rates of production of the transactinium elements in the r process, but, of the greatest importance, the triad of classical isotopes ^{232}Th, ^{238}U, and ^{235}U has been completed by two additional no less important nuclides ^{129}I and ^{244}Pu. Hohenberg, Podosek, and Reynolds (see ref. 13) have carried out careful investigations of radiogenic ^{129}Xe in meteorites and have assigned its origin to decay of now vanished ^{129}I with a half-life of 1.72×10^7 years.

The average value of the ratio $(^{129}\text{I}/^{127}\text{I})_M$ obtained by them for ten chondrites is $(1.07 \pm 0.04)\times 10^{-4}$.

More recently Podosek[14] found for the St. Severin meteorite $(^{129}\text{I}/^{127}\text{I})_M = (0.785 \pm 0.035)\times 10^{-4}$. Therefore we can reliably take $(^{129}\text{I}/^{127}\text{I})_M = (0.9 \pm 0.2)\times 10^{-4}$, and then the relation

$$\left(\frac{^{129}\text{J}}{^{127}\text{J}}\right)_M=\left(\frac{^{129}\text{I}}{^{127}\text{I}}\right)_\Delta e^{-\lambda_{129}\delta} \tag{24}$$

permits determination of the time δ of formation (cooling) of the solid bodies of the solar system, which was impossible to do previously on the basis of the long-lived isotopes of uranium and thorium[8] (see Eq. (29a) below).

However, a particularly remarkable role in study of the history of the solar system and galaxy belongs to plutonium-244.

PLUTONIUM-244 IN METEORITES, IN THE EARTH, AND IN THE MOON

Mass-spectroscopic measurements of plutonium separated from Precambrian bastnäsite confirm the presence of plutonium 244 in nature.

The history of the investigation of traces[9] of ^{244}Pu begins in 1960 when P. Kuroda of the University of Arkansas suggested that the excess of isotopes of gaseous xenon: ^{131}Xe, ^{132}Xe, ^{134}Xe, and ^{136}Xe, which was observed in the achondritic Pasamonte meteorite originates from spontaneous fission of some heavy radioactive nucleus.[16] In contrast to the Earth, meteorites should not contain primary xenon—the product of galactic nucleosynthesis, and therefore the uranium/xenon ratio in the Earth's system, including the atmosphere, should differ from the same ratio in meteorites, and the excess of stable xenon isotopes observed in meteorites can be explained only by their fission origin. Here the now extinct (decayed) radioactive nuclei—the source of the fission xenon—must have a half-life sufficiently small ($<10^8$ years) in comparison with the age of the Earth and at the same time comparable with the time of solidification of meteorites, since after implantation into meteorites they must remain there until the meteorites cool off and solidify, which assures that the gaseous fission products are captured and retained.

There can be four main sources of fission xenon: spontaneous fission of ^{238}U, neutron fission of ^{235}U, spontaneous fission of ^{244}Pu, and spontaneous fission of ^{247}Cm.

However, uranium-238 is not suitable because of the long half-life, and fission of uranium-235 by neutrons gives too small a contribution if the duration of solidification of meteorites is small in comparison with the age of the Earth. Consequently there remain only two possible sources—curium-247 and plutonium-244. The first of these has a half-life ($T_{1/2} = 1.5\times 10^7$ years) 5.5 times smaller than the second. Therefore it is natural to suggest that plutonium-244 ($T_{1/2} \approx 8.2\times 10^7$ years) is the only nuclide responsible for the presence of fission xenon in meteorites.

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[8] As Pikelner [15] recently noted, the results on the duration of synthesis and the periods of its enhancement are very sensitive to the content in meteorites of ^{244}Pu and other short-lived isotopes, which so far has not been determined with very good accuracy.

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[9] $$^{244}\text{Pu}\xrightarrow[T_{1/2}=8.2\cdot10^7\text{ y}]{\alpha}{}^{240}\text{U}\xrightarrow{\beta}{}^{240}\text{Np}\xrightarrow{\beta}{}^{240}\text{Pu}\xrightarrow{\alpha}{}^{236}\text{U}\xrightarrow{\alpha}{}^{232}\text{Th}\longrightarrow\ldots$$

In 1969 an excess of fission tracks was observed in the uranium-rich mineral whitlockite from the St. Severin meteorite (the uranium chemically accompanies plutonium); however, as before it was impossible to state with certainty that the fissioning nucleus was ^{244}Pu, since an unambiguous answer to this question can be given only by agreement of the ratio of xenon isotopes in the meteorite with the ratio of xenon fission fragments of ^{244}Pu.

Recognizing the importance of this fact, the group at the Oak Ridge National Laboratory electromagnetically separated 13 mg of plutonium-244 from large quantities of plutonium bombarded by neutrons, and after a 23-month delay, so that enough ^{244}Pu could undergo fission, they measured directly[17] the ratio of xenon fission fragments of artificial plutonium-244. This ratio turned out to be: ^{131}Xe:^{132}Xe:^{134}Xe:^{136}Xe = 25:88:92:100, which is in exact agreement with the mysterious ratio of xenon isotopes observed in meteorites. This means that plutonium-244 actually existed in the solar system at the time of formation of the meteorites.

The half-life of ^{244}Pu is 82 million years. Therefore the average concentration of ^{244}Pu on the Earth should now be only $(1/2)^{56}$ of its concentration at the time of formation of meteorites $\sim 4.6\times10^9$ years ago. Even if ^{244}Pu existed initially in the same quantity in which its decay product ^{232}Th is now distributed on the Earth (4.4×10^{-8} g/g of Earth), the upper limit of the present concentration on the Earth of ^{244}Pu must be expected to be 3×10^{-25} g/g, while even the most sensitive mass-spectroscopic measurements require $\sim 10^7$ atoms (4×10^{-15} g) of plutonium for possible detection. It is clear from this how difficult it is to detect natural plutonium-244. It would be possible to look for it only in an Earth mineral in which plutonium was selectively concentrated to the highest degree.

Plutonium-244 was found on the Earth[18] in 1971 by the Los Alamos group (D. Hoffman, F. Lawrence, J. Mewherter, and F. Rourke) in the rare earth mineral bastnäsite. The bastnäsite from which Pu-244 was extracted contains significant quantities of cerium, whose enrichment in this mineral amounts to 5×10^5 in comparison with the mean terrestrial abundance of cerium. The initial material for the investigation consisted of 260 kg of California ore which contained approximately 10% (26 kg) of bastnäsite. This is equivalent to 10.5 kg of pure CeO_2. The initial product in the form of a HDEHP solution was separated into three main parts, each containing 8.5 kg of bastnäsite. All three parts were processed separately: Tetravalent plutonium was separated from cerium and other impurities by the well known procedure for chemical separation of plutonium and was subjected to mass spectrometric analysis. For reliable identification of ^{244}Pu, indicator ions of ^{236}Pu and ^{242}Pu in accurately known amounts were added to each fraction. The first plutonium fraction show the presence of too great a quantity of Th, which was estimated in the second fraction; however, the second fraction still did not give enough ^{242}Pu indicator ions to carry out an accurate analysis. In the third fraction ^{244}Pu was found and was confirmed by two independent measurements of equal portions of the third fraction. The first portion gave a ratio of the indicator ^{242}Pu to ^{244}Pu of 284 ± 40, and the second—266 ± 40. A total of 268 ions were detected in the region of mass 244, of which 26 were assigned to experimental noise.

The ratio ^{242}Pu/^{244}Pu measured in the mass spectrometer with allowance for the known amount of ^{242}Pu indicator introduced (5.65×10^9 atoms) permits calculation of the absolute amount of ^{244}Pu recorded in the third fraction—2×10^7 atoms (8×10^{-15} g) in 8.5 kg of bastnäsite. The total error in the measurements does not exceed 30% and the final result, in the opinion of the authors, is the observation of $(2.0^{+0.3}_{-0.7})\times10^7$ atoms of ^{244}Pu in 8.5 kg of bastnäsite or 1×10^{-18} g of ^{244}Pu in 1 g of bastnäsite.[18] The possibility of introduction into the investigated sample of artificial ^{244}Pu obtained as the result of the nuclear activities of man is excluded by the low value in the same fraction of the ratio ^{239}Pu/^{244}Pu.

As noted in an editorial in Nature,[19] with the discovery at Los Alamos plutonium-244 left the category of extinct radioactivities and joined uranium-235 in the category of almost extinct radioactivities.

More recently the study[20] of lunar mountain rock obtained by Apollo 14 permits the statement that traces of plutonium-244 have now been observed also on the moon.

The xenon data obtained on heating sample 14301, which contained the mineral whitlockite, showed a significant fission component which it was possible to assign reliably to spontaneous fission of ^{244}Pu. In particular, the spectrum of xenon isotopes obtained from sample 14301 turned out to be completely identical to the spectrum from spontaneous fission of ^{244}Pu.

In addition, if we assume the same initial Pu/U ratio for the lunar rock 14301 and the Pasamonte meteorite, then we find that the rock sample 14301 was formed no later than 120 million years after the Pasamonte meteorite. The crystals of whitlockite in lunar rocks 10057, 12040, 12063, and 12064 also contain densities of fission tracks much higher than expected from the slowing down of cosmic rays or spontaneous fission of ^{238}U, but in these samples, in contrast to sample 14301, it was not possible to separate clearly the fission component from the nuclear disintegration.

If we take for the ratio ^{244}Pu/^{238}U at the time of formation of the meteorites and the moon (4.6×10^9–4.7×10^9 years ago) the value 0.015 obtained from the concentrations of fission xenon, then on the basis of the average terrestrial abundance of ^{238}U of 1.1×10^{-8} g/g it is possible to calculate the present average abundance in the solar system of ^{244}Pu—8×10^{-27} g/g. This means that the degree of enrichment of ^{244}Pu in California bastnäsite amounts to 1.3×10^8, or 130 times greater than the enrichment of cerium.10)

PLUTONIUM 244 AS A COSMOLOGICAL CHRONOMETER

Study of the content of plutonium-244 and iodine-129 in the solar system permits refinement of the picture of nucleosynthesis and the formation of the solar system.

The ^{244}Pu/^{238}U ratio was determined by Wasserburg, Huneke, and Burnett[22] in 1969 in the uranium-rich mineral whitlockite from the St. Severin meteorite $(^{244}\mathrm{Pu}/^{238}\mathrm{U})_M = 0.035 \pm 0.006$. The uniform model of

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10) Attempts to observe ^{244}Pu fission tracks in the same California bastnäsite did not yield positive results [21].

Fowler and Hoyle gives for this ratio (see Eq. (22)) the value

$$\left(\frac{^{244}\mathrm{Pu}}{^{238}\mathrm{U}}\right)_M = \left(\frac{^{244}\mathrm{Pu}}{^{238}\mathrm{U}}\right)_\Delta e^{-\lambda_{244}\delta} \tag{25}$$

$$= \frac{a_{244}}{a_{238}}\frac{\lambda_{238}}{\lambda_{244}}\frac{(1-e^{-\Delta/\lambda_{244}})}{1-e^{-\Delta/\lambda_{238}}}e^{-\lambda_{244}\delta}$$

$$= 0.010 \pm 0.002$$

for $\delta = 0.1 \times 10^9$ years. The exponential model leads to the same result.

This discrepancy of a factor of 3.5 led to Fowler to suggest the "explosion" of the sudden r process of nucleosynthesis, which occurred immediately before the formation of the solar system[11] (Fig. 5). Such an explosion is acquired in order to produce enough ^{244}Pu, since this cannot be achieved in continuous nucleosynthesis prior to formation of the solar system. However, chemical fractionation providing an enrichment of plutonium relative to uranium by roughly a factor of three in the formation of the mineral whitlockite could remove the discrepancy in the relative yield $(^{244}\mathrm{Pu}/^{238}\mathrm{U})_M$ without introducing a sudden flash of nucleosynthesis before formation of the solar system. The question is what is the true (unfractionated) value of $(^{244}\mathrm{Pu}/^{238}\mathrm{U})_M$. Podosek[23] recently found for the entire St. Severin meteorite $(^{244}\mathrm{Pu}/^{238}\mathrm{U})_M = 0.015 \pm 0.0014$. This may indicate significant fractionation in the mineral whitlockite, which is a lesser component of the entire meteorite.

In the table we have listed the parameters which are sufficiently reliable at the present time and which can be used to construct a model of nucleosynthesis.

From the data shown it is easy to see that the model of a single sudden synthesis at $t = \Delta'$ (without introducing continuous synthesis up to the formation of the solar system) leads to contradictory results.

In fact, from Eq. (21)

$$\frac{N_{A_1}}{N_{A_2}} = \frac{a_{A_1}}{a_{A_2}} e^{-(\lambda_{A_1}-\lambda_{A_2})\delta}. \tag{26}$$

Hence, for example, for ^{235}U if we take $\delta = 0.1 \times 10^9$ years we obtain the value $(^{235}\mathrm{U}/^{238}\mathrm{U})_M = 1.66$, which is five times the value observed. For $(^{244}\mathrm{Pu}/^{238}\mathrm{U})_M$ the discrepancy is even greater.

The discovery of ^{244}Pu on the Earth in detectable quantities gives an additional basis for rejecting the model of a single sudden synthesis which occurred long before the formation of the solar system (at a time $t = \Delta' \geq 6$ billion years ago).

Thus, we must assume that the r process of nucleosynthesis proceeded continuously for a long period of time up to the formation of the solar system, and in what follows we shall consider the model of continuous exponential synthesis whose moment of termination coincides with the moment of formation of the solar system[11)] (Fig. 6).

From Eq. (18) for the ratio of the yields of two elements A_1 and A_2 at the moment of formation of the meteorites we obtain

$$\left(\frac{N_{A_1}}{N_{A_2}}\right)_M = \frac{a_{A_1}}{a_{A_2}}\frac{1-\lambda_{A_2}T^*}{1-\lambda_{A_1}T^*}e^{-(\lambda_{A_1}-\lambda_{A_2})\delta}\frac{e^{-\lambda_{A_1}\Delta}-e^{-\Delta/T^*}}{e^{-\lambda_{A_2}\Delta}-e^{-\Delta/T^*}}. \tag{27}$$

First let us determine δ—the time between the formation of the solar system and the solidification of the meteorites.

For this purpose let us take two relatively short-lived nuclear chronometers: ^{244}Pu and ^{235}U.

Assuming $\lambda_{244} \gg 1/T^*$, $\lambda_{235} \gg 1/T^*$ and $e^{-\lambda_{244}\Delta} \ll e^{-\Delta}/T^*$, we obtain

$$\left(\frac{^{244}\mathrm{Pu}}{^{235}\mathrm{U}}\right)_M = \frac{a_{244}}{a_{235}} e^{-\delta(\lambda_{244}-\lambda_{235})}\frac{\lambda_{235}}{\lambda_{244}}. \tag{28}$$

Hence, using the data in the table, we find

$$\delta = (0.06 \pm 0.03) \cdot 10^9 \text{ years} \tag{29}$$

As has been noted above, the value of δ can also be determined from the data on $(^{129}\mathrm{I}/^{127}\mathrm{I})_M$.

If we take for the ratio of the production rates a_{129}/a_{127} the value given in the table, then for the uniform model[12] we have

$$\left(\frac{^{129}\mathrm{I}}{^{127}\mathrm{I}}\right)_\Delta = (5 \pm 2) \cdot 10^{-3}. \tag{29a}$$

Then from Eq. (24) we find $\delta = (0.10 \pm 0.02) \times 10^9$

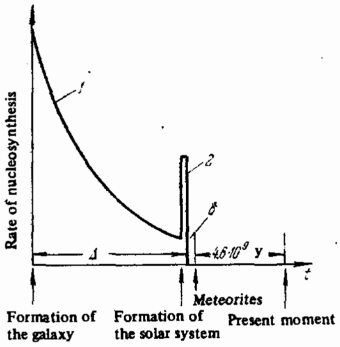


FIG. 5. Chronological model of the r process of nucleosynthesis on the assumption of a sudden flash of synthesis before formation of the solar system. 1–Exponential galactic synthesis, 2–explosion of a supernova in the region of the solar system?

	Ratio of yields		Ratio of rates of production in the r process a_{A_1}/a_{A_2} (O)
	At present (P)	At time of formation of meteorites (M) (4.6 ± 0.1) × 10⁹ years ago	
$\frac{^{244}\mathrm{Pu}}{^{238}\mathrm{U}}$	10^{-17}	0.0154 ± 0.0014	0.9 ± 0.1
$\frac{^{235}\mathrm{U}}{^{238}\mathrm{U}}$	$(7.25 \pm 0.01) \cdot 10^{-3}$	0.313 ± 0.026	1.4 ± 0.2 *)
$\frac{^{232}\mathrm{Th}}{^{238}\mathrm{U}}$	4.0 ± 0.2 **)	2.48 ± 0.15	1.65 ± 0.15
$\frac{^{129}\mathrm{J}}{^{127}\mathrm{J}}$	—	$(0.9 \pm 0.2) \cdot 10^{-4}$	1.5 ± 0.5 ***)

*With allowance for the effect of decrease in yield in the r process of odd A.

**Zartman and Wasserburg [24] found $(^{232}\mathrm{Th}/^{238}\mathrm{U})_P = 3.9 \pm 0.1$ in old North American volcanic rocks. Steiger and Wasserburg [25] found $(^{232}\mathrm{Th}/^{238}\mathrm{U})_P = 4.0$ in Canadian rocks. The value $(^{232}\mathrm{Th}/^{238}\mathrm{U})_P = 4.2$ was found in moon rocks.

***Fowler found $a_{129}/a_{127} = 1.52 \pm 0.50$, interpolating the r-process yields of ^{126}Te, ^{128}Te, and ^{130}Te.

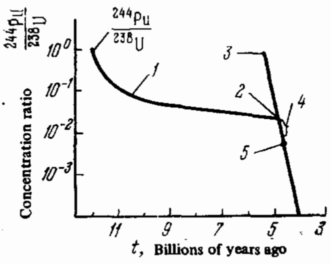


FIG. 6. Variation with time of the relative concentration ^{244}Pu/^{238}U. 1–Continuous synthesis, 2–end of nucleosynthesis, 3–sudden synthesis, 4–formation of the solar system, 5–formation of meteorites.

[11)] It is possible also to introduce an additional flash of nucleosynthesis directly at the moment of formation of the solar system; however, the relative contribution of this flash to the yield of ^{244}Pu, as Fowler has shown, is small (2.5 ± 1.5%) if we accept the data of Podosek for the St. Severin meteorite.

years. Comparing this value with the value of δ obtained from the ratio $({}^{244}\mathrm{Pu}/{}^{235}\mathrm{U})_M$, for calculation of the duration Δ and characteristic time T* we shall take δ = (0.09 ± 0.03) × 10^9 years.

From Eq. (27) we have

$$\left(\frac{{}^{235}\mathrm{U}}{{}^{238}\mathrm{U}}\right)_M=\frac{a_{235}}{a_{238}}\,\frac{1-\lambda_{238}T^*}{1-\lambda_{235}T^*}\,e^{-(\lambda_{235}-\lambda_{238})\delta}\,\frac{e^{-\lambda_{235}\Delta}-e^{-\Delta/T^*}}{e^{-\lambda_{238}\Delta}-e^{-\Delta/T^*}}, \tag{30}$$

$$\left(\frac{{}^{232}\mathrm{Th}}{{}^{238}\mathrm{U}}\right)_M=\frac{a_{232}}{a_{238}}\,\frac{1-\lambda_{238}T^*}{1-\lambda_{232}T^*}\,e^{-(\lambda_{232}-\lambda_{238})\delta}\,\frac{e^{-\lambda_{232}\Delta}-e^{-\Delta/T^*}}{e^{-\lambda_{238}\Delta}-e^{-\Delta/T^*}}.$$

Substituted the values given in Table I and δ = (0.09 ± 0.03) × 10^9 years and solving Eq. (30) for Δ and T*, we obtain Δ = 6.0 ± 2, T* = 12.0 ± 5 (in units of 10^9 years).

These quantities,

$$\begin{gathered}\delta=(0.09\pm0.03)\cdot10^9\ \mathrm{y},\qquad T^*=(12.0\pm5)\cdot10^9\ \mathrm{y},\\ \Delta=(6.0\pm2)\cdot10^9\ \mathrm{y},\quad \theta_{SS}=(4.7\pm0.1)\cdot10^9\ \mathrm{y},\\ \theta_G=(10.7\pm2)\cdot10^9\ \mathrm{y},\end{gathered}\tag{31}$$

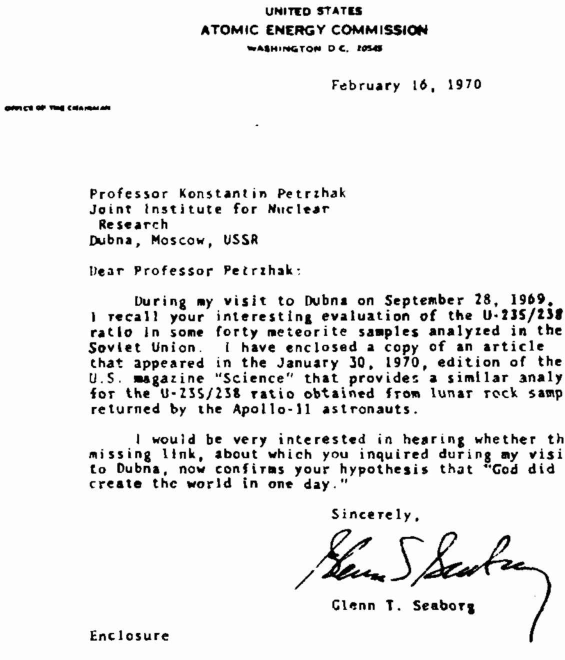
completely determine the model of nucleosynthesis.

Thus, we see that investigation of traces of plutonium-244 in meteorites, in the Earth, and on the moon permits not only improvement of the model of nucleosynthesis but also a substantial extension of the picture of the origin of the solar system. The most remarkable thing in this extension is that, in the first place, the moment of termination of nucleosynthesis, it turns out, cannot be too far removed from the moment of formation of the solar system and, in the second place, all of the bodies of the solar system (at least the meteorites, the Earth, and the moon) were formed simultaneously on the cosmological time scale in time less than or of the order of hundreds of millions of years (δ ≤ 10^8 years) (Fig. 7).

"God created the world in one day!"

PLUTONIUM-244 AND NUCLEAR STABILITY

In this chapter in connection with the discovery of plutonium-244 on the Earth in detectable quantities we discuss the possibility of variation of nuclear stability in the past as a consequence of variation of the universal constants.



UNITED STATES
ATOMIC ENERGY COMMISSION
WASHINGTON D.C. 20545

February 16, 1970

OFFICE OF THE CHAIRMAN

Professor Konstantin Petrzhak
Joint Institute for Nuclear
Research
Dubna, Moscow, USSR

Dear Professor Petrzhak:

During my visit to Dubna on September 28, 1969, I recall your interesting evaluation of the U-235/238 ratio in some forty meteorite samples analyzed in the Soviet Union. I have enclosed a copy of an article that appeared in the January 30, 1970, edition of the U.S. magazine "Science" that provides a similar analysis for the U-235/238 ratio obtained from lunar rock samples returned by the Apollo-11 astronauts.

I would be very interested in hearing whether this missing link, about which you inquired during my visit to Dubna, now confirms your hypothesis that "God did create the world in one day."

Sincerely,

Glenn T. Seaborg

Enclosure

FIG. 7

We cannot fail, however, to discuss an additional possibility of retaining ^{244}Pu on the earth in detectable quantities, although this possibility does not fit into the usual ideas of the constancy of physical laws. If we assume that the rate of radioactive decay changes with cosmological time and that Pu-244 in the past had a greater half-life, i.e., was more stable, then it would not be surprising that it was preserved until the present time.

Recently Davies[26] of Cambridge analyzed the possibility of variation of nuclear stability in the past and the effect of such variation on the qualitative aspects of the Universe.

Two-nucleon systems. The simplest nuclear system, as we know, is the deuteron. In this system each nucleon can be considered as moving in some rectangular potential well of depth V and width b. The zero kinetic energy is determined by the uncertainty principle as $\pi^2/4M_Nb^2$ (in units ħ = c = 1), where M_N is the nucleon mass. If V falls off more rapidly than $1/b^2$, the system is unbound. If $Vb^2 > \pi^2/4M_N = 5.2\times10^{-14}$ cm, then the two-nucleon system will be bound. For the triplet state of the two-nucleon system (the deuteron) we have $Vb^2 \approx 7.3\times10^{-14}$ cm and the deuteron therefore turns out to be a bound system, but rather loose. For the singlet state $Vb^2 \approx 4.5\times10^{-14}$ cm, and since the dineutron and diproton can exist only in the singlet state, they turn out to be just barely unbound.

A change in nuclear forces by a few percent may turn out to be sufficient to unbind the deuteron or, on the other hand, to bind the diproton or dineutron.

A calculation shows that a decrease in the coupling constant of the strong interaction g_S by 5% is sufficient for the binding energy of the deuteron to vanish and for it to cease to exist in nature as a stable nucleus. This would seriously change the principal nuclear reaction in stars and the entire nuclear synthesis in cosmology—the content of hydrogen in the universe would be 100%.

On the other hand, Freeman Dyson[27] pointed out the importance of the fact that the diproton is unbound. A calculation shows that an increase in g_S of only 2% would be enough to bind the diproton. If the diproton (^{2}He) were bound, there would occur in stars not the slow weak interaction $p + p \rightarrow D + e^+ + \nu$, but the fast strong interaction $p + p \rightarrow {}^2\mathrm{He} + \gamma$ and the subsequent spontaneous decay ${}^2\mathrm{He} \rightarrow {}^2\mathrm{H} + e^+ + \nu$ would not decrease the rate of burning of hydrogen with formation of diprotons, so that the entire process would occur in the form of an instantaneous explosion. Hydrogen would burn with catastrophic rapidity even in the early universe.12)

The fact that no such explosion occurred even in very remote time means that the diproton was not bound even in the earliest epoch of the universe, i.e., in this epoch nuclear forces could not have been greater than their present value by 2%. This imposes a severe limitation on the possibility of a decrease with time of the strong interaction constant g_S:

$$\left|\frac{1}{g_S^2}\frac{dg_S^2}{dt}\right| \leqslant 4\cdot10^{-12}\ \mathrm{y}^{-1} \tag{32}$$

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12)Stability of the diproton also would change nucleo synthesis in the hot model (see above). The universe would contain 100% (^{4}He + D + ^{3}He).

Many-nucleon systems. Radioactive decay. If we turn to discussion of many-nucleon systems, it turns out that study of their stability permits limits to be obtained on the change with time not only of nuclear forces but also of Coulomb repulsive forces, since the stability of heavy nuclei is determined, as is well known, by the competition of these forces.

Expressing the effect of strong interactions (nuclear forces) in terms of a surface tension Σ (the liquid-drop model) and assuming that it is proportional to g_S^4, we obtain the relation

$$\frac{Z^2}{A} < \frac{40\pi r_0^3}{3e^2}\,\Sigma \sim \frac{g_S^4}{e^2} \qquad (33)$$

(where $r_0 \approx 1.2 \times 10^{-13}$ cm, e is the electronic charge), which is the criterion of stability of a nucleus with atomic weight A and charge Ze with respect to spontaneous fission or α decay.

If e^2 is constant, the curve $Z^2/A = (g_S/g_S^{(0)})^4$ ($g_S^{(0)}$ denotes the present value of the constant g_S) separates the region of stable and unstable nuclei for various g_S. From Fig. 8 we can see that for a decrease of g_S by 25% biologically important elements such as iron become unstable. A 50% decrease in nuclear forces seriously changes the stability of even carbon.

These arguments limit the possibility of an increase with time of the constant g_S, although the models used for the evaluations can be shown to be not very rigorous.

Broulik and Trefil[28] (1971) noted that it is also possible to obtain an estimate of the limit of the possible decrease of g_S with time (or increase of e^2) if we utilize unstable nuclei not encountered in nature but which have rather large half-lives ($T_{1/2} \sim 10^7$ years). If g_S was greater (or e^2 less) in the past, there could be a time when these elements were stable against α decay. However, this time cannot be less than $\approx 10T_{1/2}$ years ago, or else a yield of these elements would be observed at present.

Broulik and Trefil selected plutonium-244, which in July 1971 was the best candidate for such an estimate since it has the longest half-life of the short-lived isotopes and had not been found on Earth at that time.

The limit obtained by them is

$$\left|\frac{1}{g_S^2}\frac{dg_S^2}{dt}\right| < 2.3\cdot 10^{-11}\ \mathrm{y}^{-1} \text{ for } \quad e^2 = \text{const},$$
$$\left|\frac{1}{e^2}\frac{de^2}{dt}\right| < 2{,}3\cdot 10^{-11}\ \mathrm{y}^{-1} \text{ for } \quad g_S^2 = \text{const}. \qquad (34)$$

However, as we know, just four months later in November 1971 ^{244}Pu was found in bastnäsite. This of course cannot be considered as an indication of a change of the constants g_S^2 or e^2, but this estimate of the limit of variation of g_S (on the basis of ^{244}Pu) now leads to an uninteresting value of $\sim 10^{-10}$ (the argument based on the nonexistence of the diproton is much stronger!).

In regard to the limits[29] of possible variation of e^2, Dyson in 1967, in studying the β decay $^{187}\mathrm{Re} \rightarrow {}^{187}\mathrm{Os}$, already obtained stricter limits

$$\frac{1}{e^2}\frac{de^2}{dt} \leqslant 3\cdot 10^{-13}\ \mathrm{y}^{-1},$$
$$\frac{1}{e^2}\frac{de^2}{dt} \geqslant -11\cdot 10^{-13}\ \mathrm{y}^{-1}, \qquad (35)$$

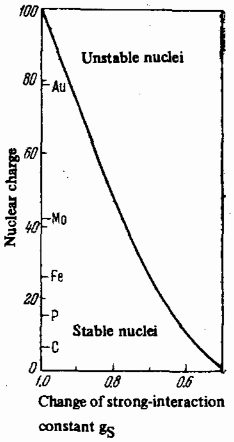

FIG. 8. Nuclear stability as a function of change in the strong-interaction constant.

These severe limits were obtained on the assumption of constancy of the strong-interaction constant: The energy difference of two nuclides ΔE was assumed to depend on time only through the constant e^2. In the first approximation, in order to estimate the dependence of ΔE on e^2 and g_S^2 at the same time, we can consider the nucleus as a sphere of radius r_0 (a Fermi gas). The Coulomb energy is obtained by averaging the proton excess in the nucleus and gives a factor proportional to Z^2e^2/r_0. The difference in the Fermi energy of the neutrons and protons gives a factor proportional to $(1/r_0^2)[(Z/A)^{2/3} - (N/A)^{2/3}]$. Assuming $r_0 \propto g_S^{-2}$, we have

$$\Delta E = A_1 g_S^4 - A_2 g_S^2 e^2. \qquad (36)$$

Thus, we see that in the case $\Delta E \approx 0$—the case of ^{187}Re and other elements, it is possible to make ΔE a constant quantity if e^2 changes in the same way as g_S^2:

$$e^2 \propto g_S^2 \sim f(t), \qquad (37)$$

where t is the time.

In Fig. 9 we have shown the limits of nuclear stability of "standard" isotopes as a function of the relative change in the strong and electromagnetic interactions. In the central part of the figure we have drawn a straight line corresponding to a variation $g_S \propto e$. The narrow crosshatched band shows the permissible region of variation of these constants. This band does not extend without limit, since on the one hand it would cross the line of stability of uranium-238 in the region of larger g_S and e, and soon after this the diproton line. On the other hand it is limited by the stability line of ^{244}Pu. In order to avoid crossing the diproton line even in the early epoch, the change of g_S must be slower, as shown by the dashed line.

Thus, nuclear stability does not forbid a simultaneous small change with cosmological time of the strong and electromagnetic interaction constants, but greatly limits this possibility.

HAS THE CONSTANCY OF THE RATE OF RADIOACTIVE DECAY WITH TIME BEEN PROVED?

Our purpose here is not to discuss the many hypotheses which assume that various fundamental physical constants actually are changing with cosmological time,[30-36] but the question arises quite naturally, especially in connection with the discovery of plutonium-244 in nature: Are there direct proofs of the constancy of the constants? One of the best known proofs involves the study of spectral lines of the remote galaxies. The set of frequencies of these lines, which arose billions of years ago, is equivalent to the set of frequencies of the same lines produced in laboratories on earth. The difference between the two sets is assigned to the Doppler red shift. This argument in favor of the constancy of the

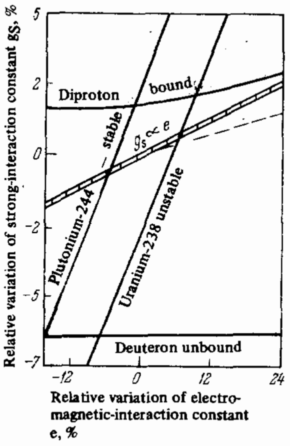

FIG. 9. Diagram of nuclear stability for change of the electromagnetic and strong-interaction constants.

constants may turn out to be even more powerful if it is confirmed that such cosmic objects as quasars are located at cosmological distances from us.

There is another proof which was obtained about 60 years ago.[37] This is the information obtained from the unique geological phenomenon occurring in certain minerals (mica, feldspar) and known by the name pleochroic halos. Pleochroic halos are colored rings which are formed around microinclusions of grains of radioactive materials as the result of emission of α particles by them. The α particles move in a straight line through the material, losing energy by ionization, the ionizing effect reaching a maximum at the end of the range and then rapidly dropping to zero.

The occurrence of the halos is due to the production of ions, and the radius of the pleochroic ring in a sheet of mica determines the range of a group of α particles of a given energy emitted by the decaying nuclei at the center of the ring. Thus, the size of the ring is direct evidence of the magnitude of the α-particle range in the past, an accurate record of events which occurred hundreds of millions of years ago. For example, if at some moment of time in the past a small grain of radioactive uranium-238 was implanted in a biotite medium, it can produce a chain of successive decays comprising in this case eight groups of α particles, each of which has a characteristic energy and consequently a characteristic range. The mica is chemically darkened at the end of the range, and over some period of time ($\sim$3 million years) eight concentric dark rings are formed in it.

The range R_α depends on the α-particle energy, and therefore a change in the energy of the α particles in cosmologically long periods of time should lead to a change in the ranges of the α particles. Furthermore if the Geiger-Nutall law $\ln R_\alpha = A + B \ln \lambda$ is valid for long periods of time, the variation of the range of the α particles can indirectly give the change in the rate of radioactive decay.

Therefore the comparison of old radii of halos with the equivalent present-day ranges can reveal any change in the decay constants λ. Such a comparison has been made by a number of authors[38,39] about 40 years ago. However, as was shown recently[40] by Richard Spector of Wayne State University, these measurements not only do not establish the constancy in time of radioactive decay constants, but in some cases even may indicate the opposite. Henderson[38] in 1934 used for measurement of the size of pleochroic rings a specially designed photometer in combination with a microscope.

Micas from Precambrian and Devonian rocks were investigated (ages from 500 to 300 million years). Here the comparison of the ranges in these ancient micas with contemporary α-particle ranges was not made directly by Henderson, but was made by conversion of the ranges in mica to the equivalent ranges in air. This required introduction of an air-mica conversion coefficient. Normalizing one range in mica to bring it into agreement with the equivalent range in air, i.e., once determining the empirical air-mica conversion factor, Henderson announced general agreement within 3% between the old and contemporary ranges for many tens of pleochroic rings of thorium and uranium. Agreement of the ranges with this accuracy at that time could be considered a convincing proof of the constancy in time of α decay, since the age of the universe was then taken as $\theta_p = 2 \times 10^9$ years and for Precambrian micas it was natural to look for a discrepancy $\Delta R/R \sim \Delta t/t \sim 25\%$ and for Devonian micas $\sim 15\%$. Now higher accuracy is necessary, since it is necessary to look for a discrepancy $\Delta R/R \sim 5\%$ for Precambrian micas and $\Delta R/R \sim 3\%$ for Devonian micas.

However, the real situation regarding the early studies of pleochroic rings is even more complicated. It turns out that Henderson did not take into account the dependence of the air-mica conversion factor on the energy of the α particles, while recent studies[41] have shown that the air-mica conversion factor changes with increase of energy in the region of α-particle energies from 4.5 MeV to 7.5 MeV by more than 7%. Allowance for this dependence, as shown by Spector,[40] leads to a deviation of several percent of the old ranges (according to Henderson's measurements) from the contemporary ranges.

It is clear from this that the early measurements of pleochroic halos can be considered a proof of the constancy of the rate of radioactive decay only with an accuracy of no better than 7–10%, whereas to check the hypothesis of variability of the constants it is necessary to have measurements whose error does not exceed 1–2% at the most.

CONCLUSION

Observation of traces of plutonium-244 and also of ^{129}I and other extinct nuclides in the solar system substantially clarifies the picture of nucleosynthesis in our galaxy. Regardless of whether the formation of the elements occurs in a continuous synthesis process right up to the origin of the solar system or with an additional flash of sudden synthesis directly at the time of formation of the solar system, the principal conclusion reduces to the fact that all nucleosynthesis occurs not in the early stages of the expansion of the universe but during an extended period of time after the formation of the galaxy. In the model of continuous exponential nucleosynthesis, as we have seen, its duration amounts to $\Delta = 6.0$ billion years

Another important conclusion regards the possibility of determining the time interval between the nucleosynthesis and the formation of the Earth and meteorites: $\delta = 10^8$ years. This value turns out to be rather small in comparison with the duration of nucleosynthesis.

Since short-lived isotopes provide useful information on the early history of the solar system, it would of course be desirable to extend the list of such chronometers as Pu-244 and I-129. Recently, for example, it was shown[42] that the isotopes ^{205}Pb ($T_{1/2} = 1.5 \times 10^7$ years) and ^{146}Sm ($T_{1/2} = 1.2 \times 10^8$ years) may turn out to be very promising chronometers (they are formed in the s and p processes of nucleosynthesis, respectively).

It is interesting also to consider the chronological possibilities of ^{247}Cm ($T_{1/2} = 1.6 \times 10^7$ years), which is formed like ^{244}Pu and ^{129}I in the r process of nucleosynthesis.[43,48]

The nucleus ^{247}Cm decays into ^{235}U, and therefore an indication of the presence of ^{247}Cm at the moment of formation of the solar system may an anomaly in the isotopic composition of uranium—an excess of uranium-235 relative to uranium-238. This excess can become significant if there is a major chemical fractionation of ^{247}Cm relative to uranium-238 in the formation of meteoritic material. Chemical fractionation substantially affects the content of actinides in the material of the solar system, and if this effect is not taken into account, the data on the yield of these elements in nucleosynthesis turn out to be uncertain.

To construct a model of nucleosynthesis it is necessary, of course, to know the true, unfractionated values of $(^{244}\mathrm{Pu}/^{238}\mathrm{U})_M$ and $(^{247}\mathrm{Cm}/^{238}\mathrm{U})_M$. In this respect the observation of U-Cm fractionation could give additional confirmation of the assumptions made in chronological investigations on the behavior of plutonium-uranium fractionation.

Podosek and Lewis[44] found for the concentration of plutonium-244 in white inclusions in the Allende meteorite the value $^{244}\mathrm{Pu}/^{238}\mathrm{U} = 0.087$, which is difficult to explain with current cosmochronological ideas (see above). Comparison of this value with the result[23] for the entire meteorite suggests an enrichment of plutonium 5–6 times in this sample relative to the unfractionated values. The chemical behavior of the actinides is such that uranium-curium fractionation is greater than uranium-plutonium fractionation, and therefore we should expect in this meteorite an appreciable anomaly in the U^{235}/U^{238} ratio.

Another important consequence in study of traces of ^{244}Pu in meteorites can be obtained if we assume that the temperature at which uranium-curium fractionation was frozen is higher than the temperature at which the meteorite retains xenon. In this case the excess ^{235}U will depend on the magnitude of the time interval between the moment of freezing of fractionation and the moment of retaining xenon, i.e., on additional details of the early history of the solar system.

Five actinide isotopes (^{232}Th, ^{235}U, ^{238}U, ^{244}Pu, and ^{247}Cm) permit information to be obtained on the production of elements in the r process in a significant mass region, and also to improve the picture of origin of the celestial bodies of the solar system on the time scale. For the latter we are indebted first of all to the study of traces of the short-lived nuclides ^{129}I and ^{244}Pu on the Earth, in the meteorites, and on the moon. However, we should also not forget that the large uncertainty in the experimental data on the content of ^{244}Pu in meteorites and the unexpected observation of this isotope on the Earth in detectable amounts still leave the possibility of explaining these data by means of the hypothesis of noninvariance of radioactive decay with respect to cosmologically long time intervals. If the rate of radioactive decay changes with cosmological time, then the picture of nucleosynthesis and the origin of the solar system is complicated significantly.[13] In order to reject this possibility experimentally, it would be desirable in particular to carry out a reinvestigation of pleochroic rings at the present level of experimental technique.

In conclusion the authors express their sincere indebtedness to Ya. B. Zel'dovich for his timely and applicable remarks and to L. E. Gurevich for extremely helpful discussions and stimulating observations. We also thank S. B. Pikel'ner for informing us of his work and K. A. Petrzhak for providing us with Glenn Seaborg's letter and for a discussion.

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[13] As Ginzburg [45] recently noted, the main question (at least from the point of view of physicists) is whether astronomy will lead to a change in any fundamental physical concepts, as desired by a number of its representatives. Examples of such changes could be the observation of changes in the physical constants with time or a deviation from well known physical laws for large densities.

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