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NUCLEAR CHRONOLOGIES FOR THE GALAXY*

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ABSTRACT

The ratios U^{235}/U^{238}, Th^{232}/U^{238}, Pu^{244}/U^{238}, and I^{129}/I^{127} have been used to obtain self-consistent solutions for the time evolution of r-process nuclei. Using $Pu^{244}/U^{238} = \frac{1}{30}$, the solutions all have a large amount of initial production, with a duration of from 0 to 10^{10} years, followed by a relatively quiescent period ($\sim 3 \times 10^9$ years) terminated by a nucleosynthetic event that possibly initiated the separation of the solar system. For values of Pu^{244} less than $\frac{1}{80}$, models of uniform synthesis terminated by a sharp nucleosynthetic event are possible.

The purpose of this Letter is to show that the relative abundances U^{235}/U^{238}, Th^{232}/U^{238}, I^{129}/I^{127}, and Pu^{244}/U^{238} at the time of formation of the solar system can provide rather severe restrictions on time scales for possible models of r-process element formation. We will further show that the existing data on the relative abundances and the relative production rates result in a very limited class of models. Previous studies have almost exclusively involved the pairs U^{235}/U^{238} and Th^{232}/U^{238} (Burbidge *et al.* 1957; Fowler and Hoyle 1960; Kohman 1961; Fowler 1962; Dicke 1969). The impetus for the present work has come from recent observations of fission products in meteoritic whitlockite produced by the decay of a transuranic element in existence at the formation of the solar system (Wasserburg, Huneke, and Burnett 1969). This isotope has been tentatively correlated with Pu^{244}. These authors point out that the ratio $Pu^{244}/U^{238} = \frac{1}{30}$ is incompatible with simple nucleosynthetic models.

The differential equation governing the abundance of isotope i with time τ is taken as $dN_i/d\tau = -\lambda_i N_i + P_i p(\tau)$, where P_i is a constant and $p(\tau)$ some function of τ; λ_i is the decay constant of species i and is zero for a stable nucleus. The integral of this equation is

$$N_i(\tau) = P_i \exp(-\lambda_i \tau) \int_0^\tau \exp(\lambda_i \xi) p(\xi) d\xi\,, \qquad N_i(0) = 0\,. \quad (1)$$

Let the termination of nucleosynthetic activity be taken as T, the time interval between the termination and the formation of planetary objects in the solar system be taken as Δ (i.e., Δ is the period of free decay), and the time of formation of planetary objects be taken as t years ago. Then the relative abundance of two isotopes at the end of Δ is

$$\frac{N_i(T+\Delta)}{N_j(T+\Delta)} = \frac{P_i \exp[-\lambda_i(\Delta+T)] \int_0^T \exp(\lambda_i \xi) p(\xi) d\xi}{P_j \exp[-\lambda_j(\Delta+T)] \int_0^T \exp(\lambda_j \xi) p(\xi) d\xi}\,. \quad (2)$$

For the isotopic ratios enumerated earlier, we have four equations which must be simultaneously satisfied by T, Δ, and $p(\tau)$.

The different isotopes have very different mean lives and thus place different constraints on equation (2). The I^{129}/I^{127} ratio places a strong upper limit on Δ that is

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virtually independent of the production ratio P_{129}/P_{127} and the choice of $p(\tau)$. The dependence of Δ on T is logarithmic and is very slowly varying (Wasserburg, Fowler, and Hoyle 1960).

Although I^{129} could possibly be formed in a process which does not produce U and Th, it is most reasonable to assume that U and Th must be made with Pu^{244}. Like I^{129}/I^{127}, the Pu^{244}/U^{238} ratio places a strong constraint on Δ; but, much more important, the abundance of U^{235} 4.6×10^9 years ago was still low enough so that the production of U^{235} associated with the necessary Pu^{244} production ($P_{235}/P_{244} = 2.2$) at times near the formation of the solar system would significantly alter the U^{235}/U^{238} ratio. The I^{129}/I^{127} and Pu^{244}/U^{238} equations in conjunction with the U^{235}/U^{238} equation constitute tight constraints on the function $p(\tau)$.

TABLE 1

VALUES OF CONSTANTS

	Standard Values	Range Explored
t	4.6×10^9 years	4.5–4.7×10^9 years
$(U^{235}/U^{238})_{now}$	1/137.8	...
$(U^{235}/U^{238})_{T+\Delta}$	0.31298	0.2884–0.3397
$(Th^{232}/U^{238})_{now}$	3.8	3.0–4.4
$(Th^{232}/U^{238})_{T+\Delta}$	2.3573	1.88–2.70
$(I^{129}/I^{127})_{T+\Delta}$	1.091×10^{-4}*	$\leq 10^{-8}$
$(Pu^{244}/U^{238})_{T+\Delta}$	$\frac{1}{30}$†	$\frac{1}{15}$–$\frac{1}{120}$

PRODUCTION RATIOS EXPLORED‡

P_{235}/P_{238}	1.44 ± 0.29	P_{232}/P_{238}	1.67 (see Th/U range)
P_{244}/P_{238}	0.654 (see fractionation)	P_{129}/P_{127}	1 (see I range)

DECAY CONSTANTS USED ($year^{-1}$)

λ_{238}	1.537×10^{-10}	λ_{235}	9.72×10^{-10}
λ_{232}	4.99×10^{-11}	λ_{129}	4.077×10^{-8}
$\lambda_{244}{}^{a}$	8.474×10^{-9}	$\lambda_{244}{}^{f}$	1.02×10^{-11}

* Hohenberg, Podosek, and Reynolds (1967).
† Wasserburg, Huneke, and Burnett (1969).
‡ Seeger, Fowler, and Clayton (1965).

The approach used was to examine various possible functional forms $p(\tau)$ for a simultaneous solution to equation (2) using the physical constants in Table 1 within the ranges shown. For a function $p(\tau,\alpha,\beta, \ldots)$ with parameters α, β, . . . , we have determined the simultaneous solutions (Δ,T) to the three equations (2) (for I^{129}/I^{127}, Pu^{244}/U^{238}, and U^{235}/U^{238}) for all values of the parameters. The Th^{232}/U^{238} equation is used as an ancillary constraint to exclude models which give ratios outside the acceptable range (3.0–4.4).

For any function $p(\tau)$ a single nucleosynthetic event corresponds to the case $T = 0$. This case is illustrated in both Figures 1*a* and 1*b*. Because of the Pu^{244}/U^{238} and I^{129}/I^{127} constraints, no solution exists within the possible range of the constants.

We then consider cases of $p(\tau) = K + S\delta(\tau) + d\delta(T - \tau)$, with $K = 0$ or 1, corresponding to a model of continuous nucleosynthesis ($K = 1$) with contributions from single events at the beginning ($S > 0$) and/or the end ($d > 0$) of nucleosynthesis.

Curves for $d = 0$ and $d = 10^9$, $S = 0$, are shown in Figure 1*a*. Note that all the curves are raised with increasing d and that the concordant solutions at points A and B for $d = 0$ with the I^{129}/I^{127} curves move upward and to the right as d increases. The large displacement of B' with increasing d is due to the concomitant addition of U^{235} with Pu^{244} (for $d = 10^9$ the intersection B' does not exist).

Point B in Figure 1*a* is a triple intersection at $\Delta = 8.37 \times 10^7$ years and $T = 7.39 \times 10^9$ years. If we demand that the Pu^{244}/U^{238} curve also pass through point B, this requires that the ratio of $\frac{1}{30}$ from the whitlockite be larger than the average solar-system value by a fractionation factor of 3.8. The complete trajectories for these two intersections are

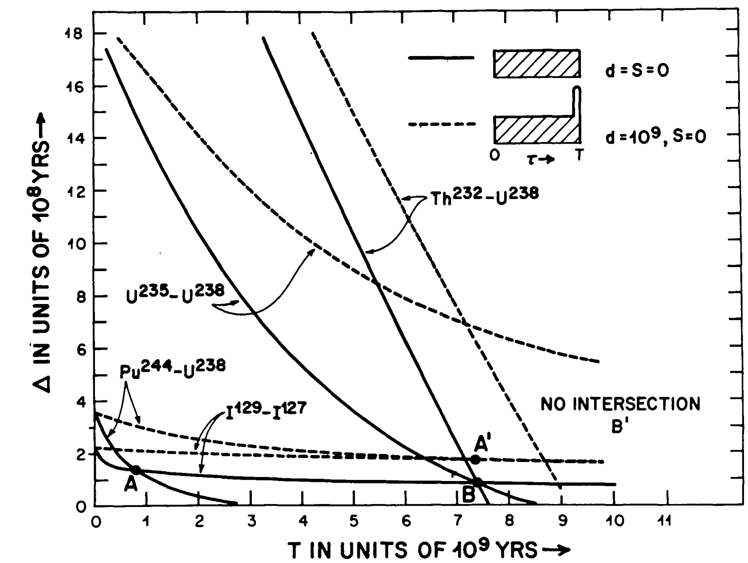

FIG. 1*a*.—(Δ, T) trajectories for each of the four isotope ratios for $p(\tau) = K + d\delta(\tau - T)$. Full curves are for uniform production (YONI model); dashed curves are for uniform production terminated by a spike. Points A and B are for concordance of Pu^{244}/U^{238} and I^{129}/I^{127} and of U^{235}/U^{238} and I^{129}/I^{127}, respectively. (Note concordance of the Th^{232}/U^{238} curve at point B as well.)

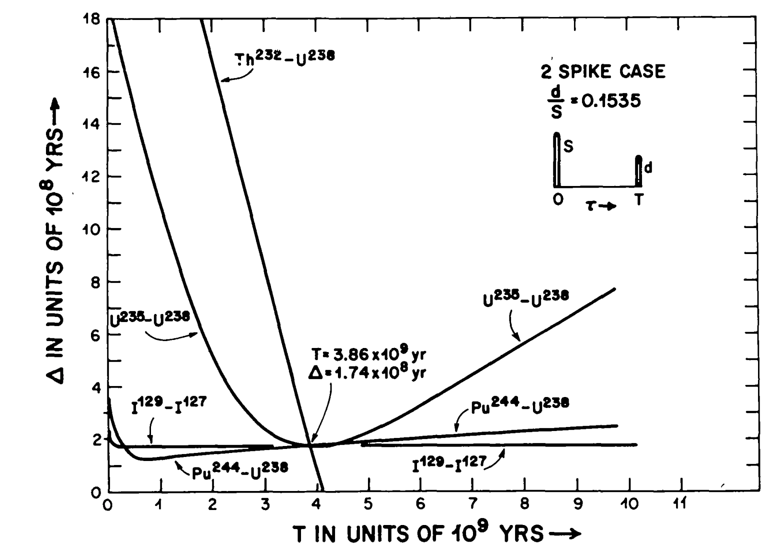

FIG. 1*b*.—Trajectories for each of the four isotope ratios for the model $p(\tau) = S\delta(\tau) + d\delta(T - \tau)$. Note the completely concordant point and the double intersection of Pu^{244}/U^{238} and I^{129}/I^{127} curves.

shown in Figure 2 for all values of d (curves *1* and A) as well as for fractionation factors of 1, 2, 3, and 4. The intersection of two curves, (U^{235}/U^{238}, I^{129}/I^{127}) and (Pu^{244}/U^{238}, I^{129}/I^{127}), is a solution to the three pertinent equations. Note that no intersection exists between curves *1* and A or curves *4* and A. Fractionation by more than 3.8 permits no solutions. Fractionations of less than 1.5 yield an unreasonable ratio $Th^{232}/U^{238} \geq 5.1$ and $T \geq 18 \times 10^9$ years. Thus any fractionation less than 1.5 or greater than 3.8 will not yield a solution. A decrease in P_{235}/P_{238} by 20 percent with a Pu^{244}/U^{238} fractionation of 1.20 will, however, yield a solution with $Th^{232}/U^{238} = 4.1$. Let us ignore the I^{129}/I^{127} constraint for this model and consider the concordance curve of Pu^{244}/U^{238} and U^{235}/U^{238}. The Th^{232}/U^{238} values along this curve are high and range from a minimum of 4.3 at $\Delta = 0$ to the maximum allowed value of 4.4 at $\Delta = 2.7 \times 10^7$ years. The fraction made in the final event, $\lambda d/(1 - e^{-\lambda T} + \lambda d)$, is 1.5 percent for a stable isotope and 14 percent for U^{235} if iodine is neglected.

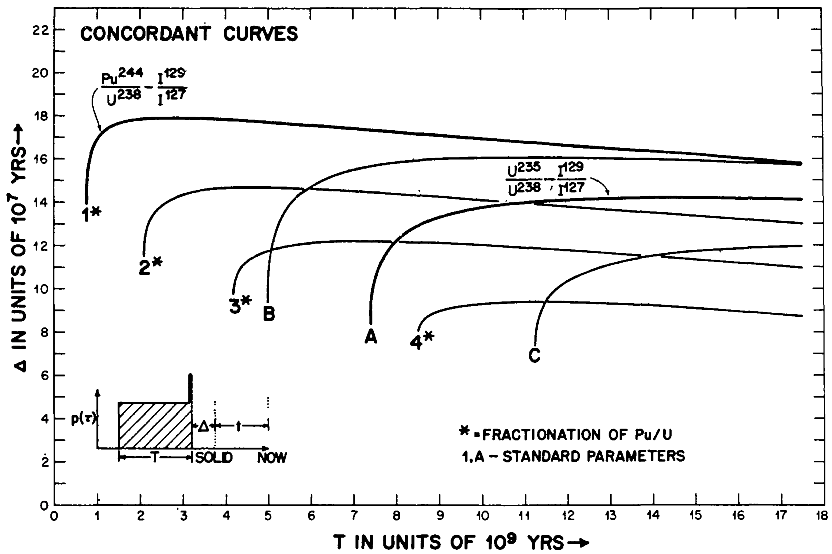

FIG. 2.—Concordant solutions of Pu^{244}/U^{238} and I^{129}/I^{127} (*1*) and concordant solutions of U^{235}/U^{238} and I^{129}/I^{127} (A) for $p(\tau) = K + d\delta(\tau - T)$. Curves *1*, *2*, *3*, *4*, are for $Pu^{244}/U^{238} = \frac{1}{30}, \frac{1}{60}, \frac{1}{90}$, and $\frac{1}{120}$, respectively. Curves B and C are for $P_{235}/P_{238} = 1.152$ and 1.728, respectively. This is equivalent to a shift in t of $\pm 2.7 \times 10^8$ years.

For the standard parameters used, there is no solution for uniform, continuous nucleosynthesis terminated by an event of arbitrary strength. A solution may be found at the extreme limits of all of the constants or more easily with Pu/U fractionation or some mechanism of nuclear destruction which preferentially destroys U^{235}. A ratio $Pu^{244}/U^{238} \leq \frac{1}{60}$ allows solutions for uniform synthesis with a spike at the end.

We now explore the case for $S \geq 0$. It is found that a concordant point for U^{235}/U^{238}, I^{129}/I^{127}, and Pu^{244}/U^{238} exists for $K = 0$, $d/S = 0.1535$, $T = 3.86 \times 10^9$ years, and $\Delta = 1.74 \times 10^8$ years, as shown in Figure 1*b*. This point yields $(Th^{232}/U^{238})_{now} = 3.8$. Thus it is a solution for all four isotope pairs. For $K = 1$, $S \geq 0$, and $d \geq 0$ the locus of triple concordance can be approximated by

$$\Delta(S/K,d/S) = 17.4 \times 10^7 - 3.36 \times 10^{-3}[T(S/K,d/S) - 3.86 \times 10^9] . \quad (3)$$

The case $K = 0$ is the low-T endpoint of the curve described by equation (3).

The ratio $(Th^{232}/U^{238})_{now}$ has been calculated along the curve and yields the values 3.8, 4.1, 4.4, 5.0, and 7.1 for $T = 3.9$, 4.9, 6.2, 8.2, and 13.4×10^9 years, respectively.

Solutions for this ratio greater than 4.4 are rejected, since they exceed the possible assigned limit. It follows that $3.86 \times 10^9 \leq T \leq 6.15 \times 10^9$ years, $S/K \geq 3.5 \times 10^{10}$ years and that the relative intensities are virtually fixed at $0.1535 \geq d/S \geq 0.124$. It also follows that the maximum contribution of continuous nucleosynthesis for a stable element is less than 14 percent. Note that solutions exist only for $d > 0$. Thus models without terminal events (e.g., Dicke 1969) do not yield solutions. Variations of the constants do not fundamentally alter the qualitative nature of the solutions presented here. It should be noted that values of $(Pu^{244}/U^{238})_{T+\Delta}$ less than $\frac{1}{30}$ yield a wider class of allowed solutions, whereas no solutions exist for $(Pu^{244}/U^{238})_{T+\Delta} \geq \frac{1}{15}$.

As an extension of this production model, we have examined the case

$$p(\tau) = \begin{cases} 1, & 0 \leq \tau \leq S, \\ k + d\delta(\tau - T), & S \leq \tau \leq T. \end{cases}$$

This model corresponds to a period of continuous, uniform nucleosynthesis for $\tau \leq S$ followed by a second period of continuous synthesis of a different intensity for $S \leq \tau \leq T$, and terminated by an event of strength d. For $k = 0$ there are compatible solutions for all equations for $S \leq 10^{10}$ years, $T - S \geq 1.5 \times 10^9$ years. There are sets of compatible solutions for both $k = 0$ and $k \neq 0$. In all these solutions $k(T - S)/[k(T - S) + S + d] \leq 0.14$. This is analogous to the other concordant models in which the dominant element production occurs in the first stage. For $k \neq 0$ the maximum fraction of stable elements made in the interval $S \leq \tau \leq T$ decreases with increasing S and is always less than that for the two-spike model $p(\tau) = K + S\delta(\tau) + d\delta(\tau - T)$.

Models with $dN_j/d\tau = -\lambda_j N_j + P_j \exp(-\omega_e \tau) + \omega_g N_j + P_j d\delta(\tau - T)$ were studied following Fowler (1962). For $d = 0$ no solutions exist for all values of ω_e and ω_g. For $\omega_g = 0$ solutions exist for $1/\omega_e < 2 \times 10^9$ years, with $\omega_e d \sim 0.1$ and $(Th^{232}/U^{238})_{now} < 4.4$.

For this investigation of a variety of production functions $p(\tau)$, using the "standard" values of the constants, we conclude that a nucleosynthetic model is required that has at least two distinct peaks separated by a time of low production. This time interval is about 3×10^9 years and is virtually model-independent. The first peak is an order of magnitude larger than the last peak or the integrated intervening continuous production. More subtle details of the production function are not determined, the initial peak possibly being a very rapid event or a long-time-scale continuous event. The time interval with low production is necessary in order for the U^{235} produced earlier to decay sufficiently so that the U^{235} produced with the Pu^{244} in the terminal event yields the required U^{235}/U^{238} ratio.

The values Δ are about $1\text{–}2 \times 10^8$ years for all the models in which the last event is represented as a delta function. A finite duration for this last event would reduce Δ. If this cannot be drastically decreased, then it is necessary that the Al^{26} (produced by a *p*-process) reported by Clarke *et al.* (1969) be produced in a later, separate event because of the short Al^{26} half-life.

The simplest interpretation may be obtained for the two-spike model. In this case the time scales are $3.86 \times 10^9 \leq T \leq 6.15 \times 10^9$ years and $1.66 \times 10^8 \leq \Delta \leq 1.74 \times 10^8$ years, the total time being $8.63 \times 10^9 \leq t + \Delta + T \leq 10.9 \times 10^9$ years. We may associate the initial event with the collapse of the proto-Galaxy and the creation of *r*-process nuclei in a phenomenon of short duration. This was followed by a low level of *r*-process element formation terminated by another sudden event, which initiated the formation of the solar system and contributed most of the Pu^{244} and I^{129} and 80 percent of the U^{235}. This type of model is in accord with the observation that the abundance of heavy elements in population I stars is independent of age. This observation has caused some workers (e.g., Unsöld 1969) to suggest that major element synthesis took place on a short time scale during the collapse of the Galaxy. If it is assumed that the astronomically observed elements may be considered as representing r-process nucleosynthesis, it would appear that these independent approaches are in good agreement.

Alternatively, we may consider the long-time-scale, steady element production with a total time $t + \Delta + T \leq 16.3 \times 10^9$ years and $S \leq 10^{10}$ years. The quiescent time is from 1.5×10^9 to less than 3.86×10^9 years. The sequence is the same as for the double-spike case, but the "initial event" is a period of uniform element synthesis.

Unless there is significant chemical fractionation of Pu, we must conclude that an event took place almost immediately prior to the formation of the solar system. This suggests that a local event (e.g., a supernova) could possibly have initiated the formation of the solar system as suggested, among other things, by Cameron (1962).

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