Reports

Radioisotopes and the History of Nucleosynthesis in the Galaxy

Abstract. *Nearly all of the heavier elements seem to have been assembled by successive neutron captures occurring in two distinct processes: the s (slow) process refers to neutron capture at a rate which is slow compared to the intervening beta-decay; the r (rapid) process refers to neutron capture at a rate which is rapid compared to the beta process. It is becoming increasingly apparent that simple models for galactic r-process nucleosynthesis are inadequate. Modern astronomical observations, which indicate that the bulk of r-process synthesis may have occurred early in the life of the galaxy, cannot be ignored. Recent data on the fissiogenic xenon in whitlockite from the St. Severin meteorite also place stringent conditions on permissible models for element synthesis. It appears that neither sudden nor continuous models for element formation are consistent with isotopic data now available. I propose a more complex model for the synthesis of solar system material in which the r-process is allowed to occur in three distinct modes: a "prompt" synthesis early in the history of the galaxy, a "continuous" synthesis whereby r-process products are continuously added to the galactic mix, and a "last-minute" synthesis which enriches the solar nebula with r-process material shortly before the formation of the solar system. Calculations based on the present abundances of uranium-235, uranium-238, and thorium-232 and the measured abundances of iodine-129 and plutonium-244 present when meteorites began to retain xenon indicate that the galactic age is between 8.0 and 8.8 billion years, with the initial "prompt" synthesis accounting for 81 to 89 percent of the total r-process material ever produced, the "last-minute" synthesis contributing between 11 and 13 percent, and 0 to 8 percent occurring in the continuous mode. The time interval between the isolation of the solar nebula from galactic r-process and the onset of xenon retention in meteorites lies between 176 and 179 million years.*

In the past, numerous attempts have been made to calculate the age of the galaxy from the decay of long-lived radioisotopes (*1–4*). As long as only a few isotopes with reasonably similar half-lives were considered, relatively simple models for nucleosynthesis could be constructed which were consistent with the isotopic data.

Precise measurements of the ratio of I^{129} (half-life, 17 million years) to stable I^{127} present when meteorites began to retain xenon (*5*) have provided further conditions which must be satisfied by allowable models for nucleosynthesis. More recently, Wasserburg *et al.* (*6*) reported an extremely high value for the ratio of Pu^{244} to U^{238} at the onset of xenon retention in the St. Severin meteorite. This result has placed even greater restrictions on the acceptable models.

I shall assume that the isotopes considered here were simultaneously produced by galactic r-process synthesis (*7*). Although it is possible that small amounts of I^{129} could be produced in the solar nebula by irradiations with high-intensity particles, there is no evidence for such an energetic solar history (*8–10*). I further assume that the fissiogenic xenon in the St. Severin meteorite results from the spontaneous fission of Pu^{244}. Detailed studies of fissiogenic xenon in the Pasamonte meteorite, including accurate measurements of the yield spectrum, virtually preclude any known source for this component other than Pu^{244} (*10, 11*). The fission spectrum for the St. Severin meteorite is identical with that found in the Pasamonte meteorite and also is only attributable to Pu^{244} (*6*). Finally I must assume that the ratio of Pu^{244} to U^{238} in the St. Severin meteorite was representative of the primitive solar system as a whole, that is, that there had been no chemical fractionation between these two elements. Within the framework of these assumptions I shall discuss various models for galactic nucleosynthesis.

First let us consider briefly a pure sudden synthesis model for r-process element formation. If nucleosynthesis were entirely confined to a short period of time, the present isotopic ratio of uranium would place this period about 6.6 billion years ago, as first pointed out by Burbidge *et al.* (*7*). However, nucleosynthesis concluding so long ago cannot account for the presence of the comparably short-lived I^{129} and Pu^{244} in the primitive solar system. Therefore it appears that the pure sudden synthesis model for galactic element formation must be ruled out.

Suppose that the r-process were occurring continuously in the galaxy and that this were the only source for r-process material in the solar system. Burbidge *et al.* suggest that certain types of supernovae may host r-process synthesis, and the ejection of this material into the interstellar medium may characterize the terminal phase of certain classes of stellar objects. Dicke (*4*) concludes that the enrichment of the interstellar medium from this process should occur at an approximately constant rate per unit mass of interstellar material. If this is true, the ratio of two radioactive isotopes, at a time t after the onset of continuous synthesis, is given by:

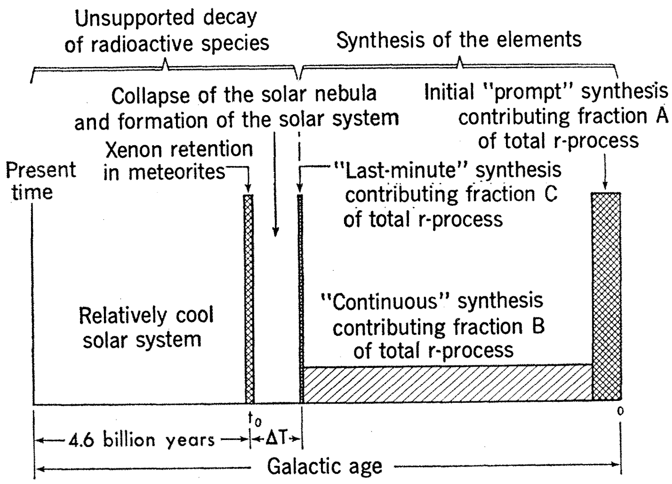
$$\left(\frac{X^i}{X^j}\right)(t) = \frac{P_i\tau_i\,(1 - e^{-t/\tau_i})}{P_j\tau_j\,(1 - e^{-t/\tau_j})}$$

where P_i and τ_i are the production rate and mean life of the isotope i.

At some time T enriched material ceased mixing with matter destined to form the solar system as a result of, say, the collapse of the solar nebula. A period ΔT followed during which the solar system formed and cooled. At this time t_0 in the primitive solar system the ratio of the two isotopes will be:

$$\left(\frac{X^i}{X^j}\right)_{t_0} = \frac{P_i\tau_i\,(1 - e^{-T/\tau_i})}{P_j\tau_j\,(1 - e^{-T/\tau_j})}\, e^{-\Delta T(1/\tau_i - 1/\tau_j)} \quad (1)$$

One important datum that should be considered at this point is derived from iodine-xenon dating of meteorites. At the time during the formation



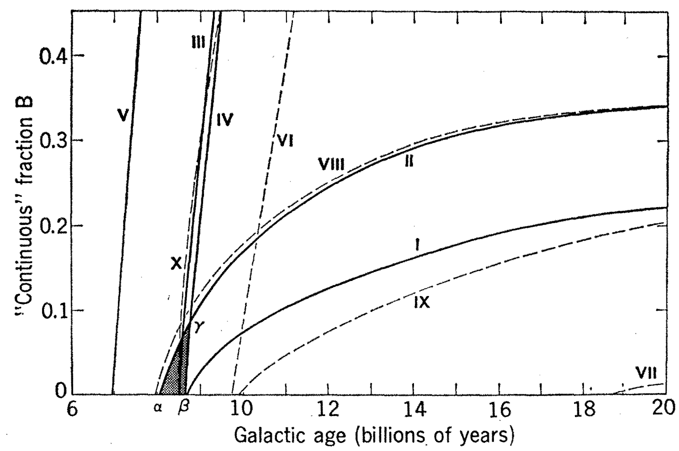


Fig. 1 (left). Model for three-mode synthesis of the elements produced by the r-process. Fig. 2 (right). Locus of points simultaneously satisfying Eqs. 2, 3, 4, 5 (curves I, II, VII, VIII, IX), Eqs. 2, 4, 5, 6 (curves III, IV, V, VI), and Eqs. 3, 4, 5, 6 (curve X). The parameters used in computing each of these curves are listed in Tables 1 and 2. On the basis of present knowledge, the parameter B and the galactic age define a point somewhere in the shaded region.

of meteorites when complete mixing of the xenon isotopes ceased, the ratio of I^{129} to stable I^{127} was $1.07 \pm 0.04 \times 10^{-4}$ (*5*). Strong evidence that this cessation of mixing occurred 4.6 billion years ago comes from the fact that the rare-gas retention ages of meteorites (as determined by the K-Ar dating method) have a rigid upper cutoff at the time of major phase separations (as determined from Pb-Pb and Rb-Sr dating methods) 4.6 billion years ago (*12*). From the data on iodine and the present isotopic ratio of uranium we can compute T and ΔT. Using 1.65 for the relative production rate P_{235}/P_{238} and 1.3 for P_{129}/P_{127} (*7*), we obtain a galactic age ($T + \Delta T + 4.6$ billion years) of 14.5 billion years and a ΔT of 83 million years.

These numbers, however, are incompatible with the ratio of Pu^{244} to U^{238} that seems to have existed in the St. Severin meteorite when isotopic mixing of xenon ceased. Wasserburg *et al.* (*6*) have reported a value of 0.035 for this ratio, with an uncertainty of about 15 percent. If we use the parameters computed above and a relative production rate P_{244} to P_{238} of 0.75 (*3*), Eq. 1 yields $0.0088\ ^{+0.0016}_{-0.0027}$ as the ratio of Pu^{244} to U^{238} expected at the cessation of isotopic mixing. It is unlikely that so large a discrepancy can be accounted for within the model since, if anything, the measured value should be a lower limit to the primitive ratio as some xenon may have been lost (*6*). It appears then that the pure continuous-synthesis model also is sorely lacking, and we must turn to more complex models for element formation.

Information that has accumulated during the last 10 years from studies of spectral lines has led to important new conclusions about the elemental composition of stars and clusters. Many astronomers believe that the new data show that the abundances of the heavy elements are remarkably uniform among the entire inventory of ordinary Population-I stars (*13, 14*); this suggests that the bulk of r-process synthesis occurred before even the oldest of the Population-I stars appeared (*4, 13*). It is therefore apparent that a modern theory of nucleosynthesis must allow for the possibility that the bulk of element synthesis was completed early in the history of the galaxy.

Table 1. Values of constants used in computations.

	Mean life
I^{129}	24.5×10^6 yr
Pu^{244}	118×10^6 yr
U^{235}	1.03×10^9 yr
U^{238}	6.51×10^9 yr
Th^{232}	20.1×10^9 yr
	Isotopic ratios
$(U^{235}/U^{238})_{now}$	0.00725
$(Th^{232}/U^{238})_{now}$	3.3 ± 0.4* 3.8 ± 0.3†
$(I^{129}/I^{127})_{t_0}$	$(1.07 \pm 0.04) \times 10^{-4}$‡
$(Pu^{244}/U^{238})_{t_0}$	0.035§
	Ratios of production rates in r-process synthesis
P_{129}/P_{127}	1.3‖
P_{232}/P_{238}	1.65 ± 0.15‖¶
P_{235}/P_{238}	1.45 ± 0.15‖ 1.65 ± 0.15¶
P_{244}/P_{238}	$0.75\ ^{+0.14}_{-0.23}$‖

* From Dicke (*4*). † From Fowler and Hoyle (*1*). ‡ From Hohenberg *et al.* (*5*). § From Wasserburg *et al.* (*6*). ‖ From Cameron (*3*). ¶ From Burbidge *et al.* (*7*).

On the other hand, we cannot exclude the possibility of some contribution by a continuous mode of nucleosynthesis. Supernova outbursts seem to be a normal feature of the galaxy, with Type-I supernovae occurring at a rate of about one per 300 years at the present time. These events, as likely sites for additional r-process synthesis, probably eject r-process material into the galactic mix at a nearly continuous rate on a cosmic time scale (*7*).

Finally, since neither continuous nucleosynthesis nor simple sudden synthesis nor a combination of the two can account for the observed Pu^{244} abundance in the primitive solar system, the postulation of a "last-minute" addition of significant amounts of r-process material seems to be necessary. This means that a complex three-mode nucleosynthesis seems to be required in order to explain isotopic data and astronomical observations now available.

Figure 1 schematically displays the three-mode model I propose. Here "prompt" synthesis is envisioned as including that element production which preceded the formation of the oldest Population-I stars. This mode may coincide with the collapse of the protogalaxy and may be limited to a time span of a few hundred million years (*4*). The "continuous" mode is due to some mechanism presently injecting enriched material into the interstellar medium, such as supernova outbursts. The "last-minute" synthesis may simply be the granularity of the continuous mode, the delta-function addition of r-process material to the solar nebula

Table 2. Values of constants used in the computation of Fig. 2.

Curve	Equations	$(Pu^{244}/U^{238})_{t_0}$	$(Th^{232}/U^{238})_{now}$	P_{235}/P_{238}	P_{244}/P_{238}	P_{232}/P_{238}
I	2,3,4,5	0.033		1.65	0.75	
II	2,3,4,5	.033		1.45	.75	
III	2,4,5,6		3.8	1.65	.75	1.65
IV	2,4,5,6		3.8	1.45	.75	1.65
V	2,4,5,6		3.3	1.65	.75	1.65
VI	2,4,5,6		3.8	1.45	.75	1.50
VII	2,3,4,5	.043		1.65	.75	
VIII	2,3,4,5	.027		1.65	.75	
IX	2,3,4,5	.047		1.45	.887	
X	3,4,5,6	.033	3.8	1.65	.75	1.65

due to a "local" supernova. Shortly after this "last-minute" addition (perhaps triggered by this event), the solar nebula collapsed and formation of the solar system began. It is assumed that, once this collapse took place with the corresponding decrease in cross section, no further contribution by galactic nucleosynthesis was made. Xenon retention by solid objects in the primitive solar system began at t_0, a time ΔT after the "last-minute" synthesis. The onset of this xenon retention occurred nearly simultaneously for a number of meteorites (*15*), approximately 4.6 billion years ago.

The ratios of I^{129} to I^{127} and Pu^{244} to U^{238}, at the time of the onset of xenon retention, and the present ratio of U^{235} to U^{238} are given by the following expressions:

$$\left(\frac{I^{129}}{I^{127}}\right)_{t_0} = \frac{P_9}{P_7} e^{-\Delta T/\tau_9} \times \left\{ Ae^{-T/\tau_9} + B\frac{\tau_9}{T}(1 - e^{-T/\tau_9}) + C \right. \quad (2)$$

$$\left(\frac{Pu^{244}}{U^{238}}\right)_{t_0} = \frac{P_4}{P_8} e^{-\Delta T(1/\tau_4 - 1/\tau_8)} \times \left\{ \frac{Ae^{-T/\tau_4} + B\frac{\tau_4}{T}(1 - e^{-T/\tau_4}) + C}{Ae^{-T/\tau_8} + B\frac{\tau_8}{T}(1 - e^{-T/\tau_8}) + C} \right\} \quad (3)$$

$$\left(\frac{U^{235}}{U^{238}}\right)_{now} = \frac{P_5}{P_8} e^{-(\Delta T + T_{ss})(1/\tau_5 - 1/\tau_8)} \times \left\{ \frac{Ae^{-T/\tau_5} + B\frac{\tau_5}{T}(1 - e^{-T/\tau_5}) + C}{Ae^{-T/\tau_8} + B\frac{\tau_8}{T}(1 - e^{-T/\tau_8}) + C} \right\} \quad (4)$$

Here T is the duration of nucleosynthesis; T_{ss} is the 4.6-billion-year age of the solar system; A, B, and C are the fractions of r-process material ever produced which result from the "prompt," "continuous," and "last-minute" modes of synthesis, respectively; P_i/P_j is the ratio of production rates in the r-process; and τ is the mean life of the isotope. Of course, there is the additional relation:

$$A + B + C = 1 \quad (5)$$

A similar expression for the present ratio of Th^{232} to U^{238} will also be useful:

$$\left(\frac{Th^{232}}{U^{238}}\right)_{now} = \frac{P_2}{P_8} e^{-(\Delta T + T_{ss})(1/\tau_2 - 1/\tau_8)} \times \left\{ \frac{Ae^{-T/\tau_2} + B\frac{\tau_2}{T}(1 - e^{-T/\tau_2}) + C}{Ae^{-T/\tau_8} + B\frac{\tau_8}{T}(1 - e^{-T/\tau_8}) + C} \right\} \quad (6)$$

The ratio of I^{129} to I^{127} at t_0 is quite well known (*5*). Determination of the amount of Pu^{244} present in a meteorite at the onset of xenon retention is made from measurements of the amount of fissiogenic xenon produced in the spontaneous fission of Pu^{244}. Precise values for the amount of Pu^{244} in the primitive meteorite are difficult to obtain since some fissiogenic xenon may have been lost over the eons. This problem does not arise in the case of I^{129} where the primitive ratio is accurately obtained through special techniques, even though the daughter product measured is also an isotope of xenon. Wasserburg *et al.* (*6*) measured the amount of fissiogenic xenon produced from the fission of Pu^{244} in the mineral whitlockite from the St. Severin meteorite. Since this measurement was made on a single mineral, rather than on an assemblage of different mineral phases, and since a high value is obtained for the plutonium-to-uranium ratio at t_0, it is thought that little xenon loss has taken place. Therefore the value of Wasserburg *et al.* is assumed for the ratio of Pu^{244} to U^{238} at t_0.

Estimations of the ratios of production rates in the r-process are available (*3, 7*); thus the question becomes: How well do Eqs. 2 through 6 determine the values of T, ΔT, A, B, and C?

Figure 2, which shows the results of these computations, displays the relationship between B and the galactic age ($T + \Delta T + T_{ss}$) resulting from the simultaneous solution of four of these five expressions. Although this was done to facilitate the calculations, the curves in Fig. 2 provide a useful display in themselves.

When I^{129}, Pu^{244}, U^{235}, and U^{238} are considered (Eqs. 2 through 5), we obtain a curve that bends sharply to the right in Fig. 2. The position of this curve depends strongly on both the ratio of production rates of U^{235} and U^{238} and on the ratio of Pu^{244} to U^{238} at the onset of xenon retention but is virtually independent of the relative production rates of iodine. I computed curves I and II by using values of 1.65 (*7*) and 1.45 (*3*), respectively, for the relative production rates of U^{235} and U^{238} and 0.035 (*6*) for the ratio of plutonium to uranium at t_0.

Using data on the isotopes of uranium, thorium, and iodine (Eqs. 2, 4, 5, and 6), I computed curves III, IV, and V. Curve V results when 3.3 is assumed for the present ratio of Th^{232} to U^{238} (*4*) and 1.65 is assumed for the relative production rates of both the pairs of nuclides U^{235}, U^{238} and Th^{232}, U^{238} (*3, 7*). Curves III and IV arise when 3.8 is chosen as the present ratio of Th^{232} and U^{238} (*1*) and values of 1.65 and 1.45, respectively, are chosen as the relative production rates of U^{235} and U^{238}. Values for these and other constants used in computing the curves of Fig. 2 are listed in Tables 1 and 2.

The intersection of these two families of curves represents a solution for Eqs. 2 through 6. This intersection will be a zone rather than a point as we must allow the various poorly known constants to vary over acceptable ranges of values.

When I recomputed curve I, using 0.027 as the value for $(Pu^{244}/U^{238})_{t_0}$, curve VIII resulted. Likewise, when 0.043 is used, curve VII results, an indication that this family of curves shifts downward (the x-intercept moving to the right) with increasing values for the ratio of Pu^{244} to U^{238} at the onset of xenon retention. However, the value 0.035 should be viewed as a lower limit for this quantity (*6*) since some xenon loss may have occurred, and curve II then represents an effective upper limit in position for this family of curves.

If the present ratio of Th^{232} to U^{238} lies between 3.3 and 3.8, then curve IV becomes a reasonable right-hand limit to the position of the second family of curves. The resulting zone of intersection, bounded by the points α, β, and γ, is the region of acceptable solutions for Eqs. 2 through 6.

Listed in Table 3 are the computed values of the parameters of nucleosyn-

Table 3. Solutions obtained at points α, β, and γ, bounding the zone of intersection in Fig. 2.

Point	Fraction of r-process synthesis occurring promptly, early in the history of the galaxy	Fraction of r-process synthesis due to continuous addition at a constant rate	Fraction of r-process synthesis contributed by a "last-minute" addition, shortly before collapse of the solar nebula	Time interval between the last event of nucleosynthesis and the onset of xenon retention in meteorites (millions of years)	Galactic age (billions of years)
α	0.87	0	0.13	179	8.0
β	.89	0	.11	176	8.7
γ	.81	0.08	.11	176	8.8

thesis and the galactic age for each of the points α, β, and γ. Within this model, the galactic age lies between 8.0 and 8.8 billion years. The interval of formation of the solar system ΔT is between 176 and 179 million years. Prompt synthesis early in the history of the solar system seems to account for the bulk (81 to 89 percent) of r-process synthesis; very little is due to the continuous mode of synthesis (between 0 and 8 percent); "last-minute" synthesis contributes from 11 to 13 percent of the total r-process material ever produced.

If the whitlockite from the St. Severin meteorite has actually suffered xenon loss, then the value 0.035 reported by Wasserburg *et al.* for the ratio of Pu^{244} to U^{238} at t_0 is an underestimate of the actual value. We can then compute the maximum value for this quantity that is still compatible with the model. Curve VI is the extreme right-hand limit for curves based on Eqs. 2, 4, 5, and 6. It assumes what are considered to be extremum values for the various parameters (Tables 1 and 2). Likewise, curve IX is computed with extremum values chosen to maximize the value of $(Pu^{244}/U^{238})_{t_0}$ and still nearly intersect curve VI. No intersection is possible for values of $(Pu^{244}/U^{238})_{t_0}$ greater than 0.047, which therefore becomes the upper limit for this ratio consistent with the three-mode model for nucleosynthesis.

An exponentially decreasing mode of nucleosynthesis has been suggested by Fowler and Hoyle (*1*), but such a model is insufficient in itself to account for the isotopic data. If the "continuous" mode of the model proposed here were exponentially decreasing in intensity, rather than constant, the basic conclusions of this report would be unaltered.

If the "last-minute" synthesis consisted of not one but two local events spaced rather closely in time, then larger values for the primitive ratio of Pu^{244} to U^{238} might be allowed. From the work of Minkowski (*16*), it is estimated that a typical supernova should enrich an area containing substantially less than 10^{-7} of the galactic mass with r-process debris before becoming sufficiently dilute to blend into the continuum. With the present rate of about one Type-I outburst every 300 years, the possibility of contamination by two nearly coincident supernovae appears to be highly remote.

One of the implied assumptions in these calculations is that the production rates in each of the three modes is the same for both iodine and the transuranic elements. If there were systematic differences in conditions and durations of individual events comprising these three modes, then differences might exist in the fractions A, B, and C for the various elements. If differences do exist, the search for them should center on iodine since this element differs greatly in mass from the other nuclides. Curve X (Fig. 2), computed from Eqs. 3 through 6, is based entirely on uranium, thorium, and plutonium. Even though data on iodine is not included, curve X intersects the zone of solutions in the figure. Furthermore, according to Dicke (*4*), stellar elemental abundances indicate that at least 60 percent of the r-process synthesis occurred early in the history of the galaxy. If true, this sets a maximum limit of about 0.3 for the "continuous" B fraction, since, with an A fraction of 60 percent, the "last-minute" C fraction is computed to be 11 percent. Therefore curve X gives a value for the galactic age between 8.5 and 9 billion years, and values for the other parameters are compatible with the previous calculations. Thus it seems that there is no apparent difficulty with the assumption that iodine is produced in constant proportion with the heavy elements during r-process synthesis in each of the three modes of element formation.

Note added in proof: Since submitting this report, I have seen a manuscript by G. J. Wasserburg, D. N. Schramm, and J. C. Huneke, to be published in *Astrophysical Journal Letters*, which treats the problem in a similar manner and arrives at basically the same conclusions.

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Moon: Infrared Studies of Surface Composition

Abstract. *Infrared reflectance studies of small lunar regions reveal several absorption bands which match those of ferrous iron in laboratory spectra of olivines and orthopyroxenes. The craters Kepler and Aristarchus exhibit absorption bands suggestive of orthopyroxene, whereas the background mare material shows a band probably due to olivine.*

I here describe observations of lunar regions approximately 7 km in diameter with scanning infrared reflectance spectrometry in the region from 0.8 to 2.2 μm. The 61-inch (155-cm)