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COSMORADIOGENIC CHRONOLOGIES OF NUCLEOSYNTHESIS*

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ABSTRACT

New methods for using radioactive decays to date the time of nucleosynthesis in our Galaxy are presented in this work. The methods depend upon a comparison of the cosmoradiogenic abundances of daughter species with the abundances of their radioactive parents. The decays which can provide useful information of this type are Re^{187}-Os^{187}, U^{235}-Pb^{207}, and U^{238}-Pb^{206} It will be shown that the amount of the enrichment of the daughter products by radioactive decay throughout the history of our Galaxy is calculable. This calculation differs from previous radioactive chronologies which have concentrated on the relative abundances of the radioactive species themselves, U^{235}, U^{238}, and Th^{232}. Definitive numerical calculations at the present time are forestalled by uncertainties in several key measurable quantities, particularly neutron-capture cross-sections and solar abundances. Tentative calculations based on probable values of these quantities indicate that galactic nucleosynthesis began at least 5×10^9 years before the formation of the solar system, and perhaps considerably earlier. Measurements capable of reducing the uncertainty of these methods are emphasized.

I. INTRODUCTION

The subject of this paper is the age of the elements, or more specifically, the time and duration of the synthesis of the neutron-rich heavy elements in our galaxy. Since these nuclear species are synthesized in catastrophic explosions, either supernovae of Type I or Type II (Burbidge, Burbidge, Fowler, and Hoyle 1957; Hoyle and Fowler 1960) or supermassive galactic explosions (Hoyle and Fowler 1963*a*), the chronology of their synthesis is equivalent to a chronology of the appropriate explosive events. The method of investigation to be employed will involve radioactive decay and theories of nucleosynthesis.

One such calculation has been made several times, most recently by Fowler and Hoyle (1960), Cameron (1962), and Dicke (1962). Those calculations are based on the fact that the relative abundances of U^{235}, U^{238}, and Th^{232} at the time of solar-system formation are known. Using the laws of exponential decay in conjunction with models of stellar activity, the time and duration of nucleosynthesis can be calculated if the relative production rates of those nuclei in supernovae are known. Unfortunately, those relative production rates are not known but must instead be calculated from theories of nucleosynthesis. Therein lies the uncertainty of that method, for the calculation must proceed in a phenomenological way by counting the number of progenitors of each nucleus and making an estimated correction for the odd-even abundance effect. Fowler and Hoyle (1960) conclude that the U^{235}/U^{238} production ratio is 1.65 ± 0.15, as is the Th^{232}/U^{238} production ratio. On the other hand, Cameron (1962) calculates a value of 1.45 ± 0.15 for the U^{235}/U^{238} production ratio and states that 1.65 is a minimum for the Th^{232}/U^{238} production ratio, which may be considerably larger. By using the values of 1.65 for both production ratios, Fowler and Hoyle compute a concordant model of nucleosynthesis which began 11.6×10^9 years ago and has decreased exponentially since that time to a present-day rate that is 26 per cent of the initial value. Cameron obtains an older commencement time by assuming an increase in the rate of nucleosynthesis shortly before the formation of the solar system. By varying these two production ratios

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over the range of values predicted by those authors, concordant models of nucleosynthesis ranging from uniform synthesis beginning about 14 billion years ago to a sudden synthesis 6.5 billion years ago may be achieved. It will be clear that any additional methods for computing the time of nucleosynthesis will be valuable in narrowing this uncertainty.

The analysis of this paper will rely upon comparing the abundances of long-lived radioactive nuclei with the cosmoradiogenic abundances of their daughters. At the time the solar system formed the abundances of both parent and daughter had some primordial value. The problem to be encountered is that of determining what percentage of the primordial-daughter abundance has resulted from radioactive decay in the time interval between the synsthesis of the radioactive parent and the formation of the solar system. Since the daughter nuclei are also produced directly by the processes of nucleosynthesis, it will be necessary to subtract the direct production from the observed abundances to obtain the cosmoradiogenic abundances. Partial solutions of this last problem represent the major contribution of this paper. Burbidge *et al.* (1957) and Fowler and Hoyle (1960) calculated the amount of cosmoradiogenic lead but were unable to make a thorough analysis of the lead problem because of insufficient data on the contributions of the slow neutron capture process.

The two decay schemes to be analyzed are the U^{235} and U^{238} decay to Pb^{207} and Pb^{206} and the long-lived beta-decay of Re^{187} to Os^{187}. Reasons for the inapplicability of the $Rb^{87}(\beta^-)Sr^{87}$ decay to this problem will also be discussed. It must be pointed out in advance that no clear-cut solution to the problem can be obtained at the present time, for the present state of knowledge of several measurable physical quantities is inadequate. It is hoped that this paper may spur future research into the specific problems that will ultimately make the solution of the chronology problem possible.

II. COSMORADIOGENIC Os^{187}

The synthesis of heavy elements occurs almost exclusively by neutron-capture chains starting with lighter elements. The essential features of these neutron-capture chains were first systematically presented by Burbidge *et al.* (1957), and have since been elaborated upon by others. The feature that remains common to all discussions of heavy-element synthesis is the need for two different processes involving neutron-capture chains: a neutron-capture chain on a slow time scale (*s*-process) that follows the line of beta stability, and rapid neutron-capture chains (*r*-process) that lead to very neutron-rich nuclei which beta-decay, after the synthesizing event, to the stable isobar with the greatest number of neutrons. Some nuclear species are produced in both processes, some in only one of the two processes, and some very rare nuclear species cannot be produced in either process.

The situation for the isotopes of Re and Os is shown in a small portion of the chart of nuclides (Fig. 1). The stable nuclear species and long-lived Re^{187} are shown by their chemical symbols, whereas unstable species are not chemically labeled. The line of beta-stability, which is the path of a neutron-capture chain for which the neutron-capture times are much greater than beta-decay lifetimes, is shown as a heavy line. This was the *s*-process path initially suggested by Burbidge *et al.* (1957). The dashed lines coming from the lower right represent isobaric beta-decays leading from the *r*-process nuclei to the stable nuclei. It can be seen that the end results of the *r*-process are, for several pertinent values of mass number, W^{184}, Re^{185}, W^{186}, Re^{187}, Os^{188}, etc. We may conclude that Re^{187} is a result of *r*-process events, whereas Os^{186} and Os^{187} are shielded from *r*-process formation.

It is by no means certain that the original *s*-process path of Burbidge *et al.* is exactly the one applicable to the bulk of *s*-process nucleosynthesis. Those authors believed that the stellar conditions for the *s*-process indicated neutron-capture times of the order of thousands of years, thereby allowing ample time for almost all beta-decays. On the other

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hand, Cameron (1959) believes that the neutron-capture times are more on the order of months, a situation that will produce considerable branching between neutron capture and beta-decay for moderately long beta-decay half-lives. We note here that our analysis is not dependent upon whether such branching occurs or not, for the same "neutron current" that passes through Os^{187} also passes through Os^{186} for any branch. For instance, any W^{185} nuclei that capture a neutron before the 76-day beta-decay occurs will follow a path through W^{186}, Re^{187}, and Os^{188}, by-passing both Os^{186} and Os^{187}. Likewise, any Re^{186} nuclei that capture a neutron before the 89-hour beta-decay can occur will also follow a path that by-passes both Os^{186} and Os^{187}. We may conclude that the synthesis of Os^{186} and Os^{187} involve initially only the *s*-process, with equal flow through Os^{186} and Os^{187}. The abundance of Os^{187} may be subsequently enriched by the decay of Re^{187}. The Re^{187} is primarily a result of *r*-process events, although it may have small contributions from branches in the *s*-process path.

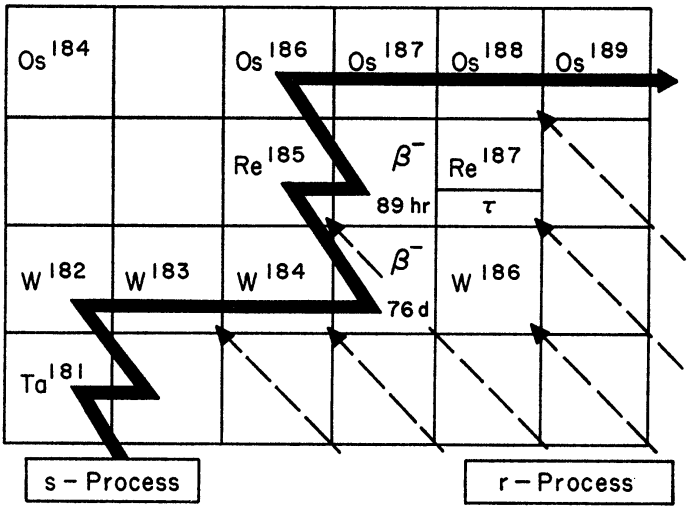

FIG. 1.—The synthesis of Re and Os. The *s*-process path, shown as the heavy line, results in the synthesis of stable species lying on the line of beta-stability. The diagonal dashed lines indicate isobaric beta-decays starting from the neutron-rich isobars synthesized in the *r*-process. The nuclei Os^{186} and Os^{187} are shielded from *r*-process nucleosynthesis by W^{186} and Re^{187}, respectively.

With the circumstances of the synthesis of the important nuclei delineated we may proceed to the special problems involved in the use of the Re^{187}-Os^{287} decay as a chronometer of nucleosynthesis.

a) *The Abundances*

The present isotopic abundance fractions for the relevant nuclei are (Strominger, Hollander, and Seaborg 1958)

$$\mathrm{Re}^{185} = 0.370\ \mathrm{Re}, \qquad \mathrm{Re}^{187} = 0.630\ \mathrm{Re}\ ;$$

$$\mathrm{Os}^{186} = 0.0159\ \mathrm{Os}, \qquad \mathrm{Os}^{187} = 0.0164\ \mathrm{Os}\ .$$

Another abundance ratio upon which the calculation will depend strongly is that of the element Os to the element Re. It will be the practice of this analysis to leave that somewhat uncertain ratio as a parameter, but some discussion of it is appropriate here.

The Re^{187}-Os^{187} decay for dating meteoritic fractionation has long been recognized as being severely handicapped by the lack of geochemical fractionation between parent and daughter elements. This handicap in determining meteoritic ages will happily be an advantage when viewed as a factor in the chronology of nucleosynthesis. Both Re and

Os are believed to be strongly siderophile (Brown and Goldberg 1949; Goldschmidt 1954). Their chalcophile tendencies are not well known, but the same Re/Os ratio is found in both the iron phase and the stone phase of the chondrite Mocs (Herr, Hoffmeister, Hirt, Geiss, and Houtermans 1961). As a result we may be justified in taking the Os/Re ratio in meteorites as being indicative of unfractionated material as far as Re and Os are concerned.

An analysis of a large number of iron meteorites reveals that the Re and Os concentrations when plotted against each other lie on a straight line with a slope Os/Re = 11.3, although the absolute concentrations vary markedly (Herr *et al.* 1961). We will take this ratio as representative of the solar system. Converting the average concentrations to an abundance on the Si = 10^6 scale gives Os = 0.601, Re = 0.0525 (Nichiporuk 1962). Only the abundance ratio is relevant to the present problem, however.

Another isotopic ratio of considerable interest is what we will take to be the primordial Os^{187}/Os^{186}-value; that is, the ratio at the time of solar-system formation. Under the reasonable assumptions that the iron meteorites are congenetic and that they initially contained primordial osmium of identical composition, the intercept of the best straight-line isochrone should give the primordial Os^{187}/Os^{186} ratio. Herr *et al.* (1961) find $Os^{187}/Os^{186} = 0.83$ at the time of formation of the iron meteorites. We will take that ratio to represent the initial value in the solar system. It will be more realistic to make the computations in terms of this primordial isotope ratio rather than the present isotope ratio to allow for the possibility that the half-life of Re^{187} in the nebular gas may differ from the half-life in meteoritic material. Noting that hereafter the chemical symbol will be used to denote abundances at the time of the formation of the solar system, we estimate

$$\begin{aligned} Os^{186} &= 0.0159\ Os, \qquad & Os^{187} &= 0.0132\ Os; \\ Re^{185} &= 0.35\ Re, \qquad & Re^{187} &= 0.65\ Re\,. \end{aligned} \tag{1}$$

The small correction to the Re composition takes account of the georadiogenic Re^{187}-decay and is made only for completeness.

Before determining the extent of the cosmoradiogenic enrichment of Os^{187}, two other small effects will be considered—the *p*-process contributions and the alteration by possible thermal neutron irradiation. The level of the *p*-process (Burbidge *et al.* 1957) in this region can be easily estimated from $Os^{184} = 0.0002$ Os. Such a small yield can be neglected in the face of other larger uncertainties.

The possible effects of thermal neutron irradiation (Fowler, Greenstein, and Hoyle 1962) are not so easy to dispose of, but the uncertainties introduced will not be large. The main effect is the enrichment of Os^{186} via thermal neutron capture by Re^{185}, whose thermal absorption cross-section is $\sigma_{th}(Re^{185}) = 104\ b$. The resulting Re^{186} has a 92 per cent decay back to Os^{186}. Smaller effects include slight changes in the abundances of other nuclei and a very small change in the Os/Re abundance ratio. Following Fowler *et al.* we may write

$$\begin{aligned} \Delta Re^{185}/Re^{185} &= -\sigma_{th}(Re^{185})\frac{n_r}{F_d\Sigma} \simeq -0.042 \\ \Delta Os^{186} &= 0.92(0.042\ Re^{185}) - \sigma_{th}(Os^{186})Os^{186}\frac{n_r}{F_d\Sigma}\,. \end{aligned} \tag{2}$$

The thermal capture cross-sections for Os^{186} and Os^{187} are not known. In order to make an estimate, we assume $\sigma_{th}(Os^{186}) \simeq 25\ b$, $\sigma_{th}(Os^{187}) \sim 100\ b$. Then

$$\Delta Os^{186}/Os^{186} \simeq 0.039\ Re^{185}/Os^{186} - 0.010 \simeq 0.067\,.$$

When other corrections are made in the same manner, we obtain two sets of values shown in Table 1.

As stated, the main effect of the thermal neutron irradiation, if it occurred, is the requirement of a lower nebular value of Os^{186}. The chronology problem will not be greatly effected by uncertainties as to which set of values to apply. Either set of numbers may be taken as representative of the solar nebula at the time of solar-system formation.

b) The s-*Process Correlation*

The determination of the amount of cosmoradiogenic enrichment of Os^{187} rests on the fact that both Os^{186} and Os^{187} are shielded from *r*-process production. After the small corrections for *p*-process formation and thermal neutron modification have been made as above, one can assert with confidence that the nebular Os^{186} is the result of *s*-process formation and that the nebular Os^{187} is the result of *s*-process production plus cosmoradiogenic Re^{187}-decay.

TABLE 1

PRIMORDIAL NEBULAR ABUNDANCES

	Without Thermal Neutrons	With Thermal Neutrons
Os^{186}	0.0159 Os	0.0148 Os
Os^{187}	0.0132 Os	0.0135 Os
Re^{185}	0.35 Re	0.35 Re
Re^{187}	0.65 Re	0.65 Re
Os/Re....	11.3	11.1

TABLE 2

SAMARIUM *s*-PROCESS CORRELATION

A	*N* (Per Cent)	σ (mb)	σN
144.	2.87	119± 55	342± 158
147.	14.94	1173±192	17600±1800
148.	11.24	258± 48	2930± 540
149	13.85	1622±297	22500±3900
150	7.36	370± 72	2770± 535
152.	26.90	411± 71	11050±1910
154..	22.84	325± 61	7440±1390

Measurements of the neutron-capture cross-sections of the isotopes of tin (Macklin, Inada, and Gibbons 1962), samarium (Macklin, Gibbons, and Inada 1963), strontium (Macklin 1963), and zirconium (Macklin 1963) have demonstrated conclusively the correctness of the *s*-process idea (Burbidge *et al.* 1957; Clayton, Fowler, Hull, and Zimmerman 1961). The case of samarium is particularly clear-cut, for it has two isotopes that can be made only in the *s*-process, Sm^{148} and Sm^{150}. The confirmation of the prediction of nucleosynthesis, that the product of abundance times the neutron-capture cross-section be equal for these two isotopes, is reproduced in Table 2. The essential equality of σN for the two *s*-only isotopes stands in marked contrast to the scattered values for the remaining isotopes. Correlations in the other elements mentioned provide similar confirmations.

The high accuracy of *s*-process correlations and the fact that Os^{186} and Os^{187} are shielded from the *r*-process allows the confident prediction

$$Os_s^{187} = [\bar{\sigma}(Os^{186})/\bar{\sigma}(Os^{187})]Os^{186} . \qquad (3)$$

The quantities $\bar{\sigma}$ are the neutron-capture cross-sections of the osmium isotopes averaged over the thermal velocity distribution appropriate to the site of *s*-process nucleosynthesis (Clayton *et al.* 1961). Unfortunately these capture cross-sections have not yet been measured. The cross-sections will be large enough to be measured with high accuracy when the separated samples are available. Since these two nuclei are far from closed nuclear-shell structures we may further anticipate that the ratio of the capture cross-sections will not be very sensitive to thermal temperature in the range $30 < kT < 60$. For this reason a measurement with moderate resolution near 30-keV neutron energy should be sufficient to obtain a good value for the cross-section ratio. Both the theory of nuclear-level densities and a systematic survey of known capture cross-sections indicate that odd-N isotopes of even-Z elements have capture cross-sections some two to four times larger than those of their even-N, even-Z neighbors. To make a tentative calculation, we will later estimate $\bar{\sigma}(\mathrm{Os}^{186})/\bar{\sigma}(\mathrm{Os}^{187}) = 0.4$. We wish to emphasize that no firm conclusion can be drawn in the absence of a measured cross-section ratio, but the correct value is expected to be sufficiently close to 0.4 to make a discussion in terms of that assumed value meaningful.

Denoting the cosmoradiogenic abundance of Os^{187} as $\mathrm{Os}_c{}^{187}$ we may write

$$\mathrm{Os}_c{}^{187} = \mathrm{Os}^{187} - \mathrm{Os}_s{}^{187} = \mathrm{Os}^{187} - [\bar{\sigma}(186)/\bar{\sigma}(187)]\mathrm{Os}^{186} . \tag{4}$$

For purposes of chronology the useful quantity is the ratio of the abundance of the radiogenic daughter to the radioactive parent.

$$\frac{\mathrm{Os}_c{}^{187}}{\mathrm{Re}^{187}} = \frac{(\mathrm{Os}^{187}/\mathrm{Os}) - [\bar{\sigma}(186)/\bar{\sigma}(187)]\,(\mathrm{Os}^{186}/\mathrm{Os})}{(\mathrm{Re}^{187}/\mathrm{Re})}\left(\frac{\mathrm{Os}}{\mathrm{Re}}\right) . \tag{5}$$

Evaluated for the two sets of primordial abundances in Table 1, this equation reduces to:

a) without thermal neutron irradiation

$$\mathrm{Os}_c{}^{187}/\mathrm{Re}^{187} = \left[0.0203 - \frac{\bar{\sigma}(186)}{\bar{\sigma}(187)}\,0.0245\right](\mathrm{Os}/\mathrm{Re}) ;$$

b) with thermal neutron irradiation

$$\mathrm{Os}_c{}^{187}/\mathrm{Re}^{187} = \left[0.0208 - \frac{\bar{\sigma}(186)}{\bar{\sigma}(187)}\,0.0228\right](\mathrm{Os}/\mathrm{Re}) .$$

These two relationships are plotted in Figure 2 for a present (Os/Re) ratio of 11.3.

If we further assume it to be most likely that $\bar{\sigma}(186)/\bar{\sigma}(187) = 0.4 \pm 0.1$ and that $\mathrm{Os}/\mathrm{Re} = 11.3 \pm 1.5$, the following tentative estimates may be made:

$$a)\ \mathrm{Os}_c{}^{187}/\mathrm{Re}^{187} = 0.120 \pm 0.031 ;$$

$$b)\ \mathrm{Os}_c{}^{187}/\mathrm{Re}^{187} = 0.130 \pm 0.030 .$$

It can be seen that there is only about an 8 per cent difference in the ratio, depending on whether thermal neutron irradiation in the amount suggested by Fowler *et al.* (1962) occurred or not. The uncertainties introduced primarily by the unknown cross-section ratio make that distinction somewhat academic at the present time. The large uncertainties can only be reduced by measurement of the neutron-capture cross-sections and by expert evaluation of the Os/Re abundance ratio.

c) *The Chronology*

A very simple picture will be used to estimate the amount of cosmoradiogenic decay as a function of the time and duration of nucleosynthesis. The origin of time, $t = 0$, is

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placed at the formation of the solar system. It is assumed that r-process nucleosynthesis (which creates the Re^{187}) began at a time T before solar-system formation and decreased exponentially as exp (Λt). (Note that t is measured backward in real time.) Then the frequency of supernovae events at the time of the solar-system formation is exp $-(\Lambda T)$ times the initial frequency in the Galaxy. With such a model, the ratio of the amount of radiogenic Os^{187} to the amount of Re^{187} at the time of solar-system formation is

$$\frac{Os_c^{187}}{Re^{187}} = \left\{ \frac{\Lambda - \lambda}{\Lambda} e^{\lambda T} \frac{1 - e^{-\Lambda T}}{1 - e^{-(\Lambda - \lambda) T}} \right\} - 1 , \qquad (6)$$

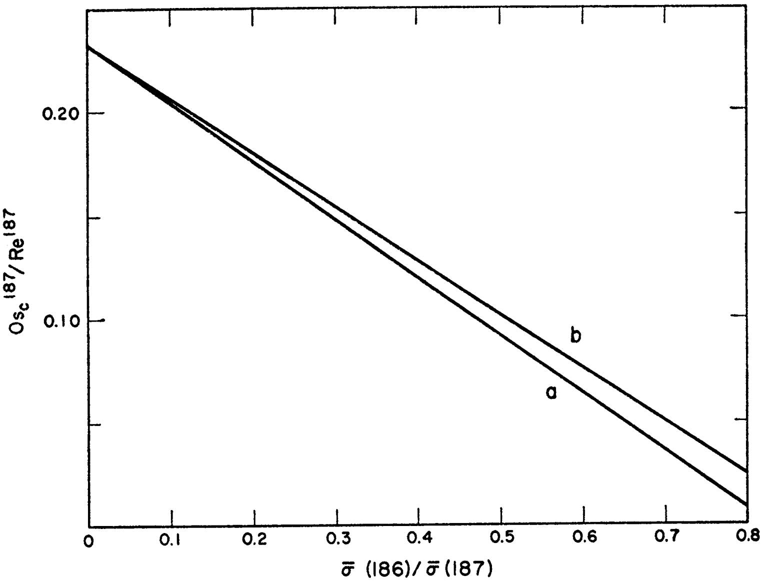

FIG. 2.—The ratio of cosmoradiogenic Os^{187} to parent Re^{187} at the time of solar-system formation as a function of the ratio of the neutron-capture cross-sections of Os^{186} and Os^{187}. From nuclear systematics one would expect $\bar{\sigma}(186)/\bar{\sigma}(187) \simeq 0.4$. Lines *a* and *b* are uncorrected and corrected, respectively, for thermal neutron irradiation of solar-system material in an amount suggested by Fowler, Greenstein, and Hoyle (1962).

where λ is the decay rate of Re^{187}. Two special cases of this result are

1. Sudden synthesis: $\Lambda \rightarrow \infty$

$$Os_c^{187}/Re^{187} = e^{\lambda T} - 1 .$$

2. Uniform synthesis: $\Lambda \rightarrow 0$

$$Os_c^{187}/Re^{187} = \frac{\lambda T}{1 - e^{-\lambda T}} - 1 .$$

The results of calculations from these formulae are shown in Figure 3. The four models shown are for sudden synthesis, uniform synthesis, and for models in which the supernovae activity at solar-system formation is 37 per cent and 14 per cent of its initial value. The amount of cosmoradiogenic decay is plotted against the time of the beginning of nucleosynthesis expressed in half-lives of Re^{187}. This method of expressing time is useful because of uncertainties in the appropriate value of the Re^{187} half-life. The horizontal dashed lines correspond to the most probable estimates of the amount of cosmoradiogenic decay for the two cases considered earlier: (*a*) $Os_c^{187}/Re^{187} = 0.12$ without correction for thermal neutron irradiation; (*b*) $Os_c^{187}/Re^{187} = 0.13$ with correction for thermal irradiation. It can be seen that the chronology estimate depends little on whether

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the correction is made or not. Recent calculations by Fowler (1963) indicate that the fraction of the primordial material exposed to the thermal neutron irradiation was much smaller than the original estimate of Fowler *et al.* (1962). In that case the correction becomes smaller than the one we have made here.

d) The Half-Life of Re^{187}

It will be clear that the Re^{187}-decay can provide a very good clock for the beginning of nucleosynthesis if the appropriate half-life can be determined. Unfortunately, this particular half-life is beset by considerable uncertainty at the present time. The best measurements of the observed beta-emission would appear to be due to Wolf and Johnston (1962), who found $\tau_{1/2} = 12 \pm 4 \times 10^{10}$ yr and a maximum decay energy of about 3 keV, and Watt and Glover (1962), who found $\tau_{1/2} \sim 3 \times 10^{10}$ yr and a maximum decay energy of 1.2 ± 0.1 keV. The discrepancy in these two measurements of the beta-emission rate is large. Other estimates of the half-life come from geological dating techniques. By using the conventional $Re^{187} = Os^{187}$ dating technique on molybdenite minerals of known age, Hirt, Tilton, Herr, and Hoffmeister (1963) conclude $\tau_{1/2} = 4.3 \pm 0.5 \times 10^{10}$ yr. In an analysis of iron meterites by the same method, Herr *et al.* (1961) conclude that their age is $(0.093 \pm 0.016)\ \tau_{1/2}$. If we tentatively assume that iron meteorites have the same age as stony meteorites, 4.4×10^9 yr, then $\tau_{1/2} = 4.7 \pm 1.0 \times 10^{10}$ yr. These last two values are in good agreement, so we will take $\tau_{1/2} = 4.5 \times 10^{10}$ yr to be the half-life of neutral Re^{187}.

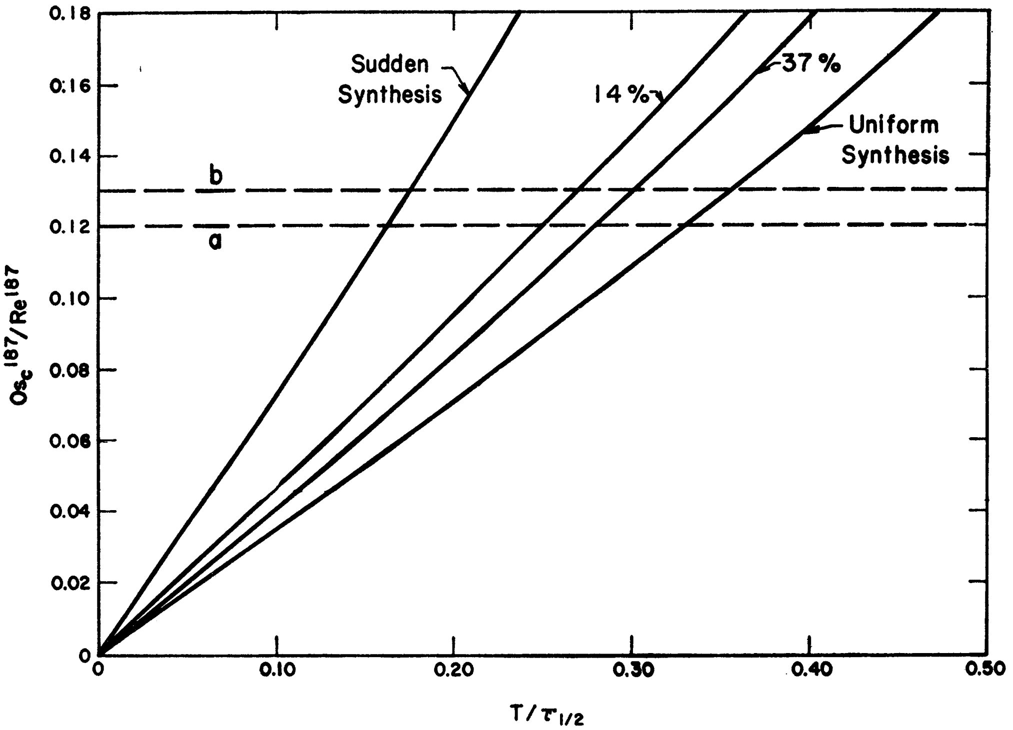

FIG. 3.—The ratio of cosmoradiogenic Os^{187} to parent Re^{187} at the time of solar-system formation calculated for various exponential models of galactic nucleosynthesis. The abscissa measures the time of the commencement of *r*-process activity in the galaxy in terms of the Re^{187} half-life, and is measured backward from the time of the formation of the solar system. The curve for each exponential model is labeled by the percentage of the initial rate of nucleosynthesis still occurring at the time of solar-system formation. The dashed lines *a* and *b* correspond to the values obtained from Fig. 2 for $\bar{\sigma}(186)/\bar{\sigma}(187) = 0.4$.

Because of the low energy of this decay, the total decay rate may contain an appreciable contribution from decay into bound final states (Gilbert 1958). If so, the half-life of the ionized atom appropriate to interstellar gas may be slightly shorter than the half-life of neutral Re^{187}. These problems are capable of solution, but for the present we will take $\tau_{1/2} = 4 \times 10^{10}$ yr to be the half-life in the interstellar medium.

From Figure 3 it can be seen that, if $\bar{\sigma}(186)/\bar{\sigma}(187) \simeq 0.4$, the interval of time between the beginning of nucleosynthesis and the formation of the solar system varies from about 0.16 $\tau_{1/2}$ for sudden synthesis to 0.33 $\tau_{1/2}$ for uniform synthesis. Taking the time of solar-system formation to be 4.6×10^9 years ago and $\tau_{1/2} = 4 \times 10^{10}$ yr, this estimate would place the beginning of galactic nucleosynthesis at a time 11×10^9 years ago for sudden synthesis and 18×10^9 years ago for uniform synthesis. Although this range of chronologies is different than that computed by Fowler and Hoyle (1960), it must be clearly understood that no disagreement really exists yet, for the present estimate is based upon quantities that are yet to be accurately determined. Most urgently needed are the appropriate neutron-capture cross-sections of Os^{186} and Os^{187} and an extensive investigation of the Re^{187}-beta-decay.

e) The Initial Heavy-Element Content of the Galaxy

The foregoing analysis has rested implicitly upon the assumption that the galactic gas initially contained no heavy elements, an assumption that has always been attractive due to its simplicity. We wish to point out at this time that the results of the Re^{187}-Os^{187} analysis may ultimately be interpreted in a different sense. If the amount of cosmoradiogenic Os^{187} determined by the measurement of the neutron-capture cross-sections indicates a much older history of nucleosynthesis than the estimates obtained from the U^{235}/U^{238} and $Th^{232}U^{238}$ production ratios, the excess cosmoradiogenic Os^{187} may be interpreted as being primordial in the galactic sense. In fact this excess Os^{187} may be identified with what the initial galactic abundance of Re^{187} would have been were it stable. A comparison of this abundance with the present Re^{187} abundance would then yield directly the fraction of *r*-process abundances initially present in the Galaxy.

An example numerical model may illustrate this point best. Let us assume that the Fowler and Hoyle chronology of nucleosynthesis, beginning 11.6×10^9 years ago and decaying exponentially to a present activity that is 26 per cent of the initial activity, is correct, and that the half-life of Re^{187} is 4×10^{10} yr. Then $T/\tau_{1/2}$ in Figure 3 is $(11.6 - 4.6)/40 = 0.17$; and the appropriate model is roughly the one for which the supernova activity at solar-system formation is 37 per cent of its initial value. The corresponding value of Os_c^{187}/Re^{187} is 0.07, which is significantly less than the anticipated value of about 0.12. The discrepancy could be explained by assuming that 5 per cent of the present *r*-process abundances were present initially in the galactic gas.

This example is intended only to be illustrative of a possible interpretation if a conflict of this type should arise. The author is by no means advocating such an interpretation. In fact, stars are known (Wallerstein, Greenstein, Parker, Helfer, and Aller 1963) for which the heavy-element concentration is less than 1 per cent of its solar value. It is worth noting, however, that this correlation does place an absolute upper limit on the initial galactic concentration of *r*-process abundances at about 10 per cent of the present concentration.

There is a way of resolving a discordance between the Re^{187}-decay and the U^{235}/U^{238} differential decay that allows one to retain the idea that the Galaxy initially contained no heavy elements. This resolution may be accomplished by adding to the average rate of galactic nucleosynthesis an intensification shortly before the formation of the solar system. The disturbance accompanying a local supernova may have provided a perturbation that could initiate the condensation of the solar nebula. This point of view has been suggested by Kohman (1961), Cameron (1962), and Murthy and Urey (1962) to account for the abundance of the extinct radioactivities Al^{26}, Pd^{107}, and I^{129}.

Possible implications of the type mentioned here must await more accurate experimental evidence on the amount of cosmoradiogenic Os^{187}. We will, however, return to this suggestion later in connection with the uranium abundance.

III. COSMORADIOGENIC Sr^{87}

It is unfortunate that the Rb^{87}-Sr^{87} decay cannot also provide a similar chronological technique. The modes of synthesis of Rb^{87} and Sr^{87} are quite analogous to the Re^{187} and Os^{187} case, and one might hope for a similar analysis of the problem. The details of the Rb^{87}-Sr^{87} correlation can probably not be solved with sufficiently high accuracy, however. The major difficulties may be summarized in the following comparisons:

1. The abundance of Re^{187} is about nine times as great as the abundance of Os^{187} that was produced directly by nucleosynthesis. Cosmic decay of 12 per cent of Re^{187} approximately doubles, therefore, the abundance of Os^{187}—an increase that can be easily measured by the σN_s correlation. The abundance of Rb^{87}, on the other hand, is approximately equal to the amount of Sr^{87} produced by nucleosynthesis. Thus the cosmoradiogenic enrichment of Sr^{87} has been close to 12 per cent—an increase that could only be accurately measured by a σN_s correlation between Sr^{86} and Sr^{87} that is accurate to something like 5 per cent.
2. The osmium isotopes lie in a mass region where the σN_s equality is essentially exact. The appropriate osmium cross-sections, furthermore, will be large enough that the ratio may be measured with high accuracy and will probably be essentially independent of temperature. The strontium isotopes, on the other hand, lie in a mass region where the σN_s product is decreasing in a manner as yet unknown. But a natural decrease of several per cent in going from Sr^{86} to Sr^{87} is quite likely, and extremely difficult to determine with assurance. The appropriate neutron-capture cross-section ratio may not be determinable to much better than 10 per cent. The proximity to the closed neutron shell at Sr^{88} causes smaller cross-sections whose effective value may vary considerably with temperature. In short, a σN_s correlation to better than 10 per cent accuracy will be quite difficult.

The question of cosmoradiogenic Sr^{87} should certainly not be ignored. Its applicability rests essentially on the following questions: Can the ratio $\bar{\sigma}(86)/\bar{\sigma}(87)$ be measured to 5 per cent accuracy? Is the ratio temperature-dependent by more than 5 per cent?

IV. COSMORADIOGENIC Pb^{206} AND Pb^{207}

The U^{235} and U^{238} produced in r-process explosions decay to Pb^{207} and Pb^{206} with mean lifetimes $\tau_{235} = 1.03 \times 10^9$ yr and $\tau_{238} = 6.51 \times 10^9$ yr. If it is possible to decide what portions of primordial Pb^{206} and Pb^{207} are due to cosmoradiogenic uranium decay, a chronology of nucleosynthesis may be calculated. The uncertainties in such an approach will be relatively large, for Pb^{206} and Pb^{207} are also synthesized directly in the s-process and in the r-process. Knowledge of the amount of cosmoradiogenic lead can only be obtained by subtracting calculated values of the amount of Pb^{206} and Pb^{207} produced directly by the s- and r-processes from the known amount of primordial Pb^{206} and Pb^{207}. The major task, therefore, of this section will be the estimation of that subtraction. The chronology calculation will also be dependent on the U/Pb abundance ratio, which is unhappily among the least well known of all abundance ratios.

This method can be seen to compare unfavorably with the Re^{187}-Os^{187} method in these respects. Radiogenic contributions aside, the Os^{186}/Os^{187} abundance ratio is determined by a simple s-process correlation, whereas the Pb^{206}/Pb^{207} abundance ratio is determined by a superposition of complicated s-process contributions with r-process contributions. The Os/Re abundance ratio is reasonably well known, whereas the U/Pb abundance ratio is not. It might be asked why this U-Pb correlation should be attempted at all since it compares unfavorably with another similar method. Among the answers to that question are these:

1. The available nuclear clues to the age of the elements are so few in number that each one must be pursued to the limit of its accuracy to obtain a self-consistent picture.
2. Since the Re^{187}-Os^{187} method is at present beset with uncertainties in the Re^{187} half-life and the neutron-capture cross-section ratio for Os^{186} and Os^{187}, its usefulness is temporarily limited.
3. The U-Pb method is intimately related to the older U^{235}-U^{238}-Th^{232} method. Many interdependent relationships between known and unknown quantities can be developed that will point the way to possible improvements in future research.

With these motives in mind, then, let us turn to the task at hand. The first requirement is a definition of the terminology that will be used. Except where explicitly noted otherwise, the chemical symbols of atoms will be used to denote their abundance at the time of solar-system formation. The subscripts s and r on the chemical symbol will, as usual, denote the abundances of the nuclear species due to the s-process and the r-process, respectively. The symbols Pb_c^{206} and Pb_c^{207} will be used to denote the contributions to the abundances of Pb^{206} and Pb^{207} from the cosmoradiogenic decay of U^{238} and U^{235} in the interval of time between nucleosynthesis and the formation of the solar system. It follows that

$$Pb^{206} = Pb_s^{206} + Pb_r^{206} + Pb_c^{206} ,$$

and

$$Pb^{207} = Pb_s^{207} + Pb_r^{207} + Pb_c^{207} . \quad (7)$$

We will further define f_s to be the fractional part of the Pb^{206} abundance due to the s-process, that is, $f_s = Pb_s^{206}/Pb^{206}$. Finally, the ratio of the production rates of U^{235} and U^{238} in r-process explosions will be denoted by R.

a) *Geophysical Quantities*

Considerable attention has been given to the problem of the isotopic composition of primordial lead. The iron meteorites are thought to be the best source of this information since it is known that they contain no uranium or thorium, whose decay since the time of formation of the meteorites would have altered the primordial composition of lead. Implicit in the foregoing statement is the belief that the iron meteorites formed at a time near the formation of the solar system itself. We conclude from the latest paper on the subject (Murthy and Patterson 1962) that the isotopic composition of primordial lead is

$$Pb^{204}/Pb = 0.020 ,$$

$$Pb^{206}/Pb = 0.189 ,$$

$$Pb^{207}/Pb = 0.206 , \quad (8)$$

and

$$Pb^{208}/Pb = 0.585 .$$

We will use these values in this paper, noting, in addition, that errors of a few per cent in these numbers could not materially affect the conclusions of this paper. The symbol Pb represents the primordial lead abundance. Its value is a few per cent less than the present solar abundance of lead.

The most accurate determination of the age of the solar system is the value $4.55 \pm 0.07 \times 10^9$ yr given by Patterson (1956) for the age of the meteorites and the earth's crust using the Pb^{207}/Pb^{206} method. Although this age does not necessarily represent the time at which the solar nebula withdrew from the products of fresh nucleosynthesis, we will take that latter time to be 4.6×10^9 years ago. We note that the methods of this paper will not be noticeably affected by arguments about the sequence of events in the few hundred million years during which the solar nebula withdrew from the interstellar gas and formed the solar system as we now know it.

The present U^{235}/U^{238} ratio is commonly accepted to be 0.00723 (Strominger *et al.* 1958). From the lifetimes of U^{235} and U^{238} it follows that the value 4.6×10^9 years ago was $U^{235}/U^{238} = 0.31$. One condition to be imposed on all models of nucleosynthesis is that the U^{235}/U^{238} abundance ratio have this value at the time of solar-system formation. The primordial uranium abundance will be denoted by its chemical symbol U. We note here that this abundance is 2.6 times the present solar abundance of uranium. It should perhaps be re-emphasized that the chemical symbols represent the primordial abundances, which are assumed to have existed 4.6 billion years ago. The present abundance in any object must take into account the georadiogenic alterations of the primordial abundances as well as the alterations of geochemical fractionation. We will not be concerned with those problems here, for the subject of this paper is the composition of the solar nebula and the history that led to it.

b) *Chronological Relationships*

The first conclusion we wish to demonstrate is the following: If the cosmoradiogenic contributions to the abundances of Pb^{206} and Pb^{207} and the present abundance of uranium are known, both the time of the beginning of nucleosynthesis and the appropriate exponential model are determined. This determination is implicitly contained in the following three relationships:

1. The total amount of U^{235} ever injected into the interstellar gas that eventually became the solar nebula can be computed at the time of solar-system formation as the sum of the remaining U^{235} and the cosmoradiogenic Pb^{207}. A similar relationship clearly holds for U^{238} vis-à-vis Pb^{208}. Since the production rate of U^{235} in supernovae is defined to be R times the production rate of U^{238}, it follows that

$$R = \frac{\mathrm{Pb}_c^{207} + \mathrm{U}^{235}}{\mathrm{Pb}_c^{206} + \mathrm{U}^{238}} . \tag{9}$$

A knowledge of these abundances therefore allows determination of the r-process production-ratio R. This relationship is quite general in the sense that it does not depend on any assumption about the time variation of the rate of nucleosynthesis. The validity of this equation requires only that there have been no chemical fractionation of lead and uranium in the interstellar gas nor in the formation of the solar system and that the production ratio be constant from event to event. The latter assumption will not have been exactly fulfilled. However, the supernovae conditions necessary for the synthesis of uranium are sufficiently well defined that only small variations in R can be envisioned. This equation applies, therefore, to an average value of R which we hereafter assume to have been constant.

2. If we invoke the exponential models of nucleosynthesis, the requirement that the U^{235}/U^{238} abundance ratio be equal to 0.31 at the time of solar-system formation determines a relationship between T (the time of the commencement of galactic nucleosynthesis), Λ (the exponential-decay constant in the rate of nucleosynthesis), and R. Explicitly this relationship is

$$\frac{\mathrm{U}^{235}}{\mathrm{U}^{238}} = 0.31 = R\,\frac{\Lambda - \lambda_{238}}{\Lambda - \lambda_{235}} \cdot \frac{\exp[(\Lambda - \lambda_{235})T] - 1}{\exp[(\Lambda - \lambda_{238})T] - 1} . \tag{10}$$

Figure 4 shows the values of R required to satisfy this expression as a function of the time for several exponential models. In these models the parameter Λ is chosen such that the rate of nucleosynthesis at the time of solar-system formation is 100 (uniform synthesis), 50, 25, 10, 2, and 0 per cent (sudden synthesis) of the rate at the commencement of nucleosynthesis.

This relationship has been used by others (Fowler and Hoyle 1960; Cameron 1962) to compute a chronological model of nucleosynthesis on the basis of values of R calculated from r-process theory.

3. For the same models the amount of cosmoradiogenic lead may be computed from the equation

$$R\frac{Pb_c^{206}}{Pb_c^{207}}=\frac{1-\{\Lambda\exp[(\Lambda-\lambda_{238})T]-\Lambda\}/\{(\Lambda-\lambda_{238})[\exp(\Lambda T)-1]\}}{1-\{\Lambda\exp[(\Lambda-\lambda_{235})T]-\Lambda\}/\{(\Lambda-\lambda_{235})[\exp(\Lambda T)-1]\}}. \tag{11}$$

This relationship is shown in Figure 5 for the same models that were shown in Figure 4.

These three relationships derived from the simple properties of exponential decay serve as the basis for the subsequent analysis.

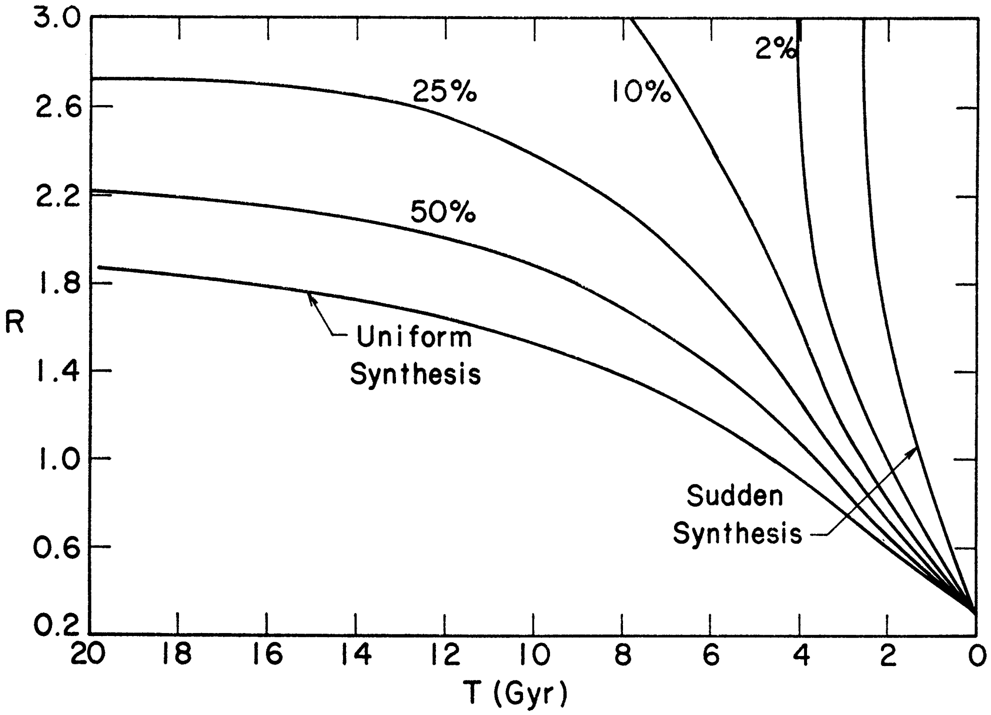

FIG. 4.—The value of R required to produce the value $U^{235}/U^{238} = 0.31$ at solar-system formation as a function of the time of the beginning of galactic nucleosynthesis. R is the production ratio of U^{235}/U^{238} in r-process explosions; its value will be assumed to be in the range $1.4 < R < 1.8$. The relationship is demonstrated for several exponential models of the rate of galactic nucleosynthesis, each curve being labeled by the percentage of the initial rate of nucleosynthesis still occurring at the time of solar-system formation.

c) The Determination of Cosmoradiogenic Lead

The primordial abundances of Pb^{206} and Pb^{207} were separated into the contributions due to the three distinct sources of those abundances in equation (7). We know in addition that $Pb^{206} = 0.189$ Pb and $Pb^{207} = 0.206$ Pb. In solving for the radiogenic contributions by subtraction, it will be convenient to divide throughout by the lead abundance:

$$\begin{aligned} Pb_c^{206}/Pb &= 0.189 - Pb_s^{206}/Pb - Pb_r^{206}/Pb \\ Pb_c^{207}/Pb &= 0.206 - Pb_s^{207}/Pb - Pb_r^{207}/Pb\,. \end{aligned} \tag{12}$$

It is from the analysis of the final two terms in each of these equations that we may ultimately obtain usable results. The following subsections contain that necessary digression.

1. *The* s-*process contributions.*—The heaviest stable nuclear species produced by

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neutron capture on a slow time scale (*s*-process) is Bi^{209}. Subsequent neutron captures form radioactive alpha-emitters which decay rapidly to the lead isotopes. In their analysis of the approach to equilibrium isotopic abundances under those circumstances, Clayton *et al.* (1961) used an incorrect energy level scheme for Bi^{210}. Clayton (1963) has pointed out that the correct equilibrium *s*-process abundances are those such that

$$\bar{\sigma}(206)\ Pb_s^{206} = \bar{\sigma}(207)\ Pb_s^{207} = \bar{\sigma}(208)\ Pb_s^{208}\,, \tag{13}$$

where $\bar{\sigma}$ is the neutron-capture cross-section of each isotope averaged over a Maxwell-Boltzmann velocity distribution with $kT \sim 20 - 60$ keV. This very simple correlation of *s*-process abundance with neutron-capture cross-section is valid only for equilibrium,

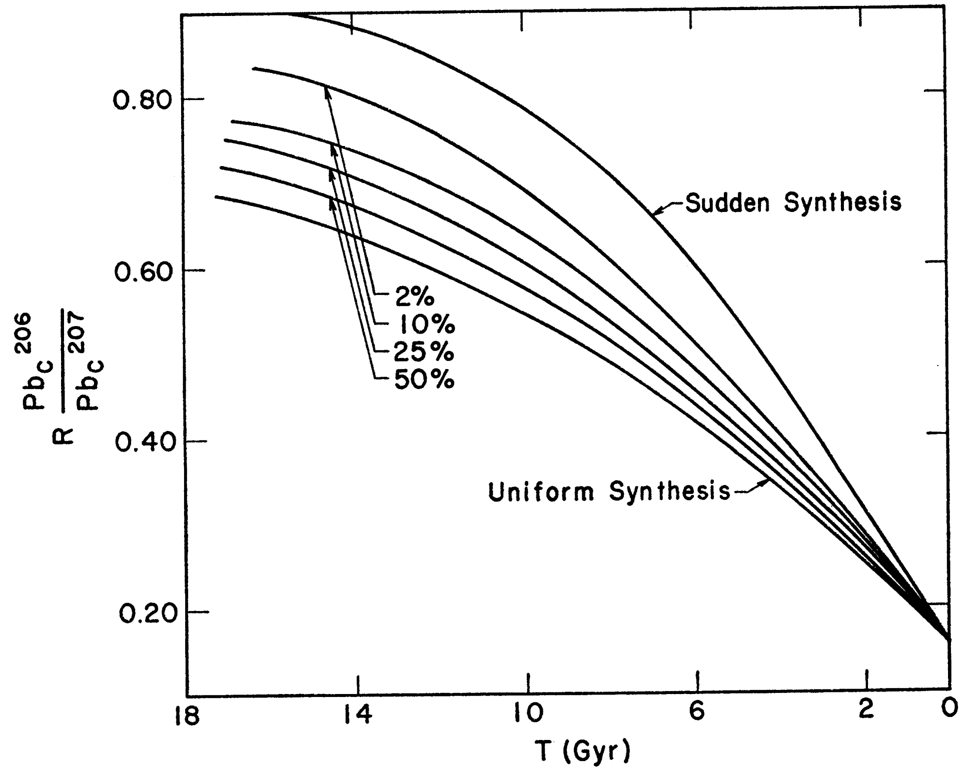

FIG. 5.—The product of *R* and the ratio of the enrichments of Pb^{206} and Pb^{207} by galactic uranium decay. The quantities are calculated for the same exponential models of nucleosynthesis as in Fig. 4.

however, whereas Clayton *et al.* (1961) showed that it is possible to have rapidly decreasing σN_s products through the lead region for a neutron flux that was just sufficient to create small amounts of lead. If this latter alternative were the case, it would necessarily be true that $\bar{\sigma}(208)\ Pb_s^{208} \ll \bar{\sigma}(206)\ Pb_s^{206}$. To eliminate this possibility, we need only examine the primordial isotope ratios,

$$Pb^{206} : Pb^{207} : Pb^{208} = 0.189 : 0.206 : 0.585\,.$$

Without going into the details here, we note that both *r*-process production and cosmoradiogenic decay contribute more strongly to Pb^{206} and Pb^{207} than to Pb^{208}. It is clear, therefore, that almost all of the Pb^{208} abundance is due to the *s*-process. If we anticipate that about one-third to two-thirds of the Pb^{206} is due to the *s*-process, it can be seen that the *s*-process abundance ratio will lie in the range

$$Pb_s^{206}/Pb_s^{208} \simeq 0.1 \text{ to } 0.2\,.$$

This ratio is equal to a reasonable estimate for $\bar{\sigma}(208)/\bar{\sigma}(206)$, so the σN_s curve cannot have dropped very much between Pb^{206} and Pb^{208}. The s-process theory further indicates that even if the σN_s curve is decreasing, the percentage decrease at $A = 207$ relative to $A = 206$ is much less than the percentage decrease at $A = 208$ relative to $A = 206$. Since the σN_s curve cannot increase in the lead region, we conclude

$$\frac{\bar{\sigma}(207)\mathrm{Pb}_s^{207}}{\bar{\sigma}(206)\mathrm{Pb}_s^{206}} = 0.9 \pm 0.1 . \tag{14}$$

Macklin (1963) has measured $\sigma(206)$ and $\sigma(207)$ as a function of neutron energy and performed integrals over the Maxwell-Boltzmann velocity distribution to obtain the average cross-sections at a given temperature. He finds that $\bar{\sigma}(206)/\bar{\sigma}(207)$ varies smoothly between 0.64 and 0.56 as kT varies from 25 to 45 keV. We may conclude that

$$\mathrm{Pb}_s^{207}/\mathrm{Pb}_s^{206} = 0.5 \pm 0.1 . \tag{15}$$

We have defined f_s as the fraction of the Pb^{206} abundance due to the s-process; $f_s = \mathrm{Pb}_s^{206}/\mathrm{Pb}^{206}$. Therefore

$$\mathrm{Pb}_s^{206}/\mathrm{Pb} = 0.189 f_s, \quad \text{and}$$

$$\mathrm{Pb}_s^{207}/\mathrm{Pb} = (0.5 \pm 0.1)\ \mathrm{Pb}_s^{206}/\mathrm{Pb} = (0.095 \pm 0.019) f_s .$$

The equations for the cosmoradiogenic contributions may then be written as

$$\begin{aligned} \mathrm{Pb}_c^{206}/\mathrm{Pb} &= 0.189\ (1 - f_s) - \mathrm{Pb}_r^{206}/\mathrm{Pb} \\ \mathrm{Pb}_c^{207}/\mathrm{Pb} &= 0.206 - (0.095 \pm 0.019)\ f_s - \mathrm{Pb}_r^{207}/\mathrm{Pb} . \end{aligned} \tag{16}$$

The parameter f_s will prove to be a useful one in the analysis of this problem. Unfortunately, its numerical value cannot be determined at present with certainty. The simplest anticipated technique for the determination rests on an accurate measurement of the Pb^{208} neutron-capture cross-section near 30 keV. About four-fifths of the Pb^{208} abundance must be due to the s-process, or $\mathrm{Pb}_s^{208}/\mathrm{Pb} \sim 0.47$. Using s-process equilibrium, $\bar{\sigma}(206)\ \mathrm{Pb}_s^{206} = \bar{\sigma}(208)\ \mathrm{Pb}_s^{208}$, we estimate

$$f_s = \frac{\mathrm{Pb}_s^{206}}{\mathrm{Pb}^{206}} = \frac{1}{0.189}\,\frac{\bar{\sigma}(208)}{\bar{\sigma}(206)}\,\frac{\mathrm{Pb}_s^{208}}{\mathrm{Pb}} \sim 2.5\,\frac{\bar{\sigma}(208)}{\bar{\sigma}(206)} . \tag{17}$$

The $\bar{\sigma}(206)$ value decreases smoothly from 6.5 mb for $kT = 20$ keV to 3.7 mb for $kT = 50$ keV (Macklin 1963). Most of the neutron flux responsible for the s-process is probably released by the $Ne^{22}(\alpha, n)Mg^{25}$ reaction in the helium-burning cores or shells of giant stars, for which $kT \sim 30$ keV and $\bar{\sigma}(206) \sim 5$ mb. To good approximation, therefore,

$$f_s \simeq 0.5\ \bar{\sigma}(208) ,$$

where $\bar{\sigma}(208)$ is the average Pb^{208} neutron-capture cross-section in mb for $kT = 30$ keV.

The Pb^{208} cross-section in this energy range is yet to be measured. However, Gibbons, Macklin, Miller, and Neiler (1961) report that the average cross-section near 30 keV for the element lead in its naturally occurring isotopic ratio is 3 ± 3 mb. This value is so much less than the cross-sections for the light lead isotopes that $\bar{\sigma}(208)$ may be inferred to be 1 mb or less. Further inferences about the value of $\bar{\sigma}(208)$ may be made from the Bi^{209} cross-section, which should be something like one to two times larger than $\bar{\sigma}(208)$. Gibbons *et al.* (1961) report $\bar{\sigma}(209) = 1 \pm 4$ mb, and Booth, Ball, and MacGregor (1958) report $\bar{\sigma}(209) = 1.8 \pm 0.7$ mb. It appears likely that $\bar{\sigma}(208)$ lies somewhere between 0.5 and 1.0 mb. Assuming that range to be correct, we may anticipate that

$$f_s = 0.4 \pm 0.1 .$$

Future cross-section measurements of high precision will allow a satisfactory determination of f_s to be made. At present we will allow f_s to be a phenomenological paramete with a probable value lying in the range indicated above.

It might be argued that the s-process cannot have produced equilibrium abundance in the lead region since the σN_s products for lead isotopes are considerably lower tha the σN_s products for atomic weights less than 200. The largest masses for which th σN_s product is unambiguously determined are Sm^{148} and Sm^{150}. Using the abundanc Sm = 0.25 (Schmitt and Smith 1962) and the cross-sections $\sigma(148) = 258$ mb an $\sigma(150) = 370$ mb (Macklin *et al.* 1963), we find that $\sigma N_s = 7.3$ for $A \sim 150$. Clayto *et al.* (1961) showed that the σN_s product should be essentially constant for $150 < A < 200$. Some confirmation of the correctness of this conclusion may be obtained fro the abundances of s-only isotopes whose cross-sections have not been measured. Usin the abundances mentioned later, Os = 0.60 and Pt = 0.89, leads to the abundanc $Pt^{192} = 0.0070$ and $Os^{186} = 0.0095$ for these two s-only isotopes. Since these isotope may be expected to have neutron-capture cross-sections of about 500 mb, the σN product may be about 5, indicating a gradual drop in σN_s of about 30 per cent as A increases from 150 to 190. Such a smooth slow decrease is to be anticipated from the wor of Clayton *et al.*

On the other hand, if $\sigma(206) = 5.3$ mb, $f_s = 0.4$, and Pb = 2.5 (Helliwell 1961), w find $\sigma(206)\,Pb_s^{206} = (5.3) \times (0.19) = 1.0$, a factor of 5 smaller than the σN_s curve nea $A = 200$. Does this not mean that the σN_s curve is dropping rapidly in the lead isotop region, and that the equilibrium s-process abundances used in the analysis of this pape are incorrect? We think not. Any neutron flux that can produce significant amounts o lead requires only a small flux to drive the lead isotopes to an equilibrium configuration It is statistically improbable that the neutron fluxes of the s-process events could hav been just sufficient to synthesize lead before they are exhausted.

A much more realistic solution to the low σN_s abundances for lead lies in a carefu evaluation of the findings of Clayton *et al.* (1961). Suppose we consider the σN_s produc per initial iron seed nucleus, a product those authors called ψ_A, as a function of the average number of neutron captures per initial iron nucleus, n_c. For a large range in n_c, sa from 80 to 130 neutrons per iron seed, ψ_A has a value near 1.5 for A less than 200, and i quite small in the lead region. On the other hand, values of $n_c > 150$ drive lead t equilibrium abundances, with a value of ψ, however, that is only 0.5. Thus the functio ψ is intrinsically small in the lead region due to the low values of the lead cross-sections With this fact in mind, we can make a more precise quantitative statement about th low values of σN_s in the lead region. After Clayton *et al.*, we let $g(n_c)$ represent the number of iron seed nuclei that have been subjected to a neutron flux that has resulted in n neutron captures per initial iron nucleus in the range dn_c. Then

$$\sigma_A N_A = \int g(n_c)\psi_A(n_c)dn_c\,. \qquad (18$$

If we now make the simple assumption that $g(n_c)$ is constant for $50 < n_c < 200$, we fin not only that the lead isotopes approximate their equilibrium distribution, but tha $\sigma_A N_A$ is a factor of about 5 higher for $A < 200$ than it is in the lead region. It is on th basis of this conclusion that we may justify taking near-equilibrium s-process abundance in the lead region even though the σN_s level has dropped markedly from its value fo $A < 200$.

2. *The* r-*process contributions.*—The extent to which the r-process events contribute directly to the abundances of Pb^{206} and Pb^{207} is difficult to determine accurately. The uncertainties are inherent in all r-process calculations and result from our inadequat knowledge of the detailed properties of nuclei lying far to the neutron-rich side of th valley of beta-stability. It is this same difficulty that has plagued the calculation of th production ratio, R, of U^{235} to U^{238} (Burbidge *et al.* 1957; Fowler and Hoyle 1960 Cameron 1962). In fact the problem of calculating Pb_r^{206} and Pb_r^{207} is quite similar to th

problem of the uranium isotopes. Not only are mass numbers 206 and 207 synthesized in the *r*-process, but short-lived trans-bismuth nuclei which rapidly decay to Pb^{206} and Pb^{207} are synthesized as well. Thus Pb_r^{206} is the sum total of *r*-process synthesis at mass numbers 206, 210, 214, 218, 222, 226, 230, and 234. Likewise, Pb_r^{207} is the sum total of *r*-process synthesis at mass numbers 207, 211, 215, 219, 223, 227, and 231. Larger mass numbers in these 4*n*-chains result in the production of U^{238} and U^{235}, respectively. It can be seen that the number of progenitors of Pb_r^{206} outnumbers those of Pb_r^{207} by the ratio 8 to 7. There is, moreover, a general tendency for even-*A* nuclei to be synthesized in the *r*-process in amounts that average some 10–20 per cent greater than those of the adjacent odd-*A* nuclei. On these very general grounds alone we would anticipate that

$$Pb_r^{206}/Pb_r^{207} \sim (8/7) \times (1.15) \simeq 1.3 .$$

By relying on the results of other workers, we may obtain a quantitative estimate of Pb_r^{206} and Pb_r^{207}. Burbidge *et al.* (1957) have calculated, on the basis of extrapolated nuclear systematics, the abundances expected from the *r*-process at each value of atomic weight on the scale Si = 10^6. Their results for the appropriate progenitors of the lead isotopes are shown in Table 3. These sums give a Pb_r^{206}/Pb_r^{207} ratio that is remarkably close to the value estimated above on general considerations; that is, $Pb_r^{206}/Pb_r^{207} = 0.477/0.369 = 1.29$. These sums, moreover, give not only the ratio Pb_r^{206}/Pb_r^{207}, but the absolute amount of each expected on the abundance scale Si = 10^6 as well.

TABLE 3

CALCULATED *r*-PROCESS YIELDS FOR PROGENITORS OF Pb^{206} AND Pb^{207}*

A	N_r	*A*	N_r
206	0.056	207	0.056
210	.037	211	.037
214	.050	215	.035
218	.048	219	.048
222	.034	223	.034
226	.045	227	.032
230	.123	231	0.127
234	0.084		
Sum	0.477	Sum	0.369

* Calculations of Burbidge *et al.* (1957).

These absolute abundances must be reinterpreted with care, however. What Burbidge *et al.* calculated was the shape of the *r*-process abundance curve. To obtain the absolute abundances requires normalization of the calculated shape to some key abundances that are known to be *r*-process products. A logical indicator of the normalization is the abundances in the so-called *r*-process peaks. Burbidge *et al.* used the abundances of Suess and Urey (1956) in this normalization process. More recent measurements of *r*-process abundances, however, have shown that normalization to the Suess and Urey abundance table results in *r*-process abundances that are something like a factor of 2–3 too large throughout the entire region of heavy atomic weights. Because of the importance of a suitable normalization of *r*-process abundances in what will follow, we digress into a somewhat detailed consideration of this problem.

The 82-neutron *r*-process peak is responsible for the present abundances of Te^{128} and Te^{130}. In normalizing their calculated *r*-process peak Burbidge *et al.* used the Suess and Urey abundance Te = 4.67. Aller (1961) estimated a better value to be Te = 3.55.

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Neutron-activation analysis of chondrites indicates even lower values; Te = 0.73 (Schindewolf 1960), Te = 0.60 in bronzite-hypersthene chondrites, and Te = 2.7 in enstatite chondrites (Goles and Anders 1962). Clayton *et al.* (1961) adopted Te = 1.83, a value that will be employed here also, thereby setting the isotopic abundances as $Te^{128} = 0.58$, $Te^{130} = 0.63$. The initial normalization of Burbidge *et al.* was chosen such that the calculated *r*-process abundances were $Te_r^{128} = 1.48$, $Te_r^{130} = 1.73$. Our adopted values of Te^{128} and Te^{130} are smaller than the latter values by factors of 0.39 and 0.36, respectively, a fact which would indicate that the calculated *r*-process abundances at atomic weights intermediate to Te, and the next *r*-process peak (Os, Ir, Pt) should also be reduced by a similar factor. The rare-earth elements lie in this region, and in light of the fact that their abundances have been determined in chondrites by neutron-activation analysis (Schmitt and Smith 1962), it will be instructive to compare observed *r*-process abundances with the calculated abundances (Becker and Fowler 1959) in this region. Table 4 shows for each value of atomic weight the observed *r*-process abundances (corrected where necessary for *s*-process contributions), the calculated *r*-process abundances of Becker and Fowler, and the ratio of observed to calculated values.

TABLE 4

COMPARISON OF OBSERVED AND CALCULATED *r*-PROCESS ABUNDANCES IN THE RARE-EARTH REGION

A	Observed N_r	Calculated N_r*	Obs N_r/Calc N_r
147	0.032	0.172	0 19
148	.042	172	.24
149	030	106	.29
150	041	106	.39
151	.046	106	44
152	.048	172	.28
153	.041	172	.24
154	.056	172	.33
155	.047	114	41
156	054	114	47
157	050	114	43
158	070	182	.38
159	.045	182	.25
160	.079	182	43
161	.068	124	.54
162	.080	.124	64
163	.090	124	72
164	110	184	60
165	.072	184	.39
166	050	184	.27
167	045	115	.39
168	.037	115	.33
169	.031	.115	.27
170	.029	.164	18
171	.021	164	13
172	.022	.164	13
173	.025	.068	.37
174	.040	.068	.59
175	032	.068	47
176	0.024	0.068	0.35
Sum	1.457	4.108	0.35

* Calculations from Becker and Fowler 1959.

We first note from Table 4 that the sum of the observed N_r is 0.35 times the sum of the calculated N_r. This ratio confirms the expectation of the previous paragraph that the calculated *r*-process abundances must be reduced by a factor approximately equal to the necessary reduction for tellurium. We conclude that the *r*-process calculations of Burbidge *et al.* have the proper shape but are, in this mass region, too large a by factor of 2.5 to 3. The ratio of the observed N_r to the calculated N_r for individual values of atomic weight varies between the extremes of 0.13 and 0.72, but the great majority of the ratios have values between 0.24 and 0.47. One intriguing aspect of these ratios is that they do not vary in a random manner at all; quite to the contrary, they exhibit correlated trends, as shown in Figure 6. The periodic peaks in this curve almost surely indicate some periodic systematic nuclear property that was not included in the formulation of Burbidge *et al.* It is the suggestion here that the rare-earth region will prove ultimately to be the one for which the most reliable N_r curve can be experimentally obtained over a large region of atomic weight. Its detailed structure will give valuable information for the construction of a more exact *r*-process theory.

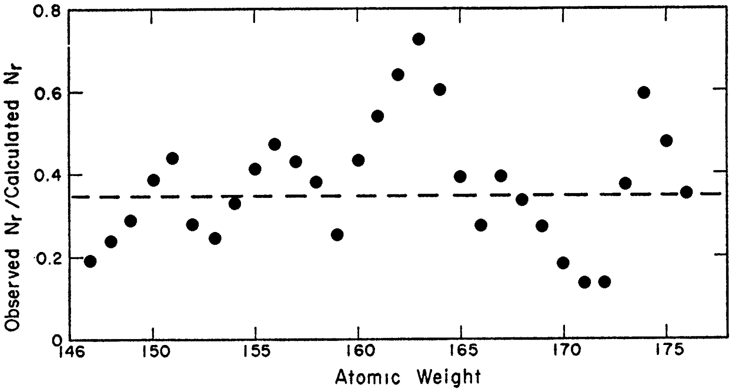

FIG. 6.—The ratio of the observed *r*-process abundances to the calculated *r*-process abundances for atomic weights in the rare-earth region. The values plotted are taken from Table 4; the observed abundances result from *s*-process subtractions from the measurements of Schmitt and Smith (1962), whereas the calculated abundances are from Becker and Fowler (1959). The horizontal dashed line shows the average normalization parameter for the *r*-process calculations in this region, $r = 0.35$.

We appear at this juncture to be somewhat far removed from the problem of calculating Pb_r^{206} and Pb_r^{207}. That appearance is deceptive, for the trans-bismuth atomic weights which are the progenitors of Pb_r^{206} and Pb_r^{207} lie in a valley beyond the second *r*-process peak. We may anticipate that the calculations of Burbidge *et al.* will reproduce the trans-bismuth abundances with roughly the same accuracy with which they reproduced the rare-earth abundances. In light of the oscillatory accuracy shown in Figure 6, we may seriously question any individual calculated abundance, which we have seen may be in error by a factor of 2. On the other hand, Pb_r^{206} is the sum of eight progenitors and Pb_r^{207} is the sum of seven progenitors. The oscillatory and random errors may be expected to largely cancel in making such a sizable sum. This belief is precisely the basis for confidence in the calculated production ratio of U^{235}/U^{238} used by other workers.

We may test this belief on the rare-earth *r*-process abundances of Table 4. To do this we will sum seven terms in a $4n$-chain starting with $A = 147$, and eight terms in a $4n$-chain starting with $A = 148$. The results are shown in Table 5. We note first that the ratios of the observed sums to the calculated sums are quite close to the average over the entire region, 0.35. This result confirms the hope that the oscillatory fluctuations in the individual ratios average out in taking partial sums of this number of terms. We may form yet another interesting conclusion from the partial sums of Table 5. It has been concluded from Table 3 that the abundance sum of the eight progenitors of Pb_r^{206} will exceed the abundance sum of the seven progenitors of Pb_r^{207} by a factor 1.29. We see in Table 5 that the partial sum of eight terms starting with $A = 148$ exceeds the partial sum of seven terms starting with $A = 147$ by the factors 1.31 and 1.20 for the observed and calculated sums, respectively. From this confirmation we conclude that the ratio $\mathrm{Pb}_r^{206}/\mathrm{Pb}_r^{207} = 1.29$ has good accuracy.

There still remains the question of exactly how to normalize the values of Pb_r^{206} and Pb_r^{207} obtained from the calculations of Burbidge *et al.* as shown in Table 3. We will write $\mathrm{Pb}_r^{206} = 0.477\, r$ and $\mathrm{Pb}_r^{207} = 0.369\, r$, where r will be a normalization parameter. It would be tempting to choose $r \simeq 0.35$ on the basis of the foregoing; however, the trans-bismuth elements lie in the r-process valley following the 126-neutron r-process peak at osmium, iridium, and platinum. Since the normalization of the calculated r-process abundances to the 126-neutron abundance peak need not be exactly the same as the normalization to the 82-neutron abundance peak, we need to compare the observed r-process abundances of Os, Ir, and Pt to the calculated values. The abundances of these three elements are yet somewhat uncertain. From neutron activation of chondrites we have the value Os = 0.6 (Bate 1960), which agrees well with a value obtained from iron meteorites, Os = 0.60 (Herr *et al.* 1961; averages performed by W. Nichiporuk). For Ir and Pt we take the values of Nichiporuk and Brown (1962), Ir = 0.31 and Pt = 0.89, based on iron meteorites. Ehmann (1961) has found a similar value in chondrites, Ir = 0.32. We estimate the s-process contributions to these three elements to be $\mathrm{Os}_s = 0.13$, $\mathrm{Ir}_s = 0.02$, and $\mathrm{Pt}_s = 0.12$. By subtraction the dominant r-process abundances of these three elements are $\mathrm{Os}_r = 0.47$, $\mathrm{Ir}_r = 0.29$, $\mathrm{Pt}_r = 0.77$, whereas the unrenormalized calculations of Burbidge *et al.* give $\mathrm{Os}_r = 1.12$, $\mathrm{Ir}_r = 0.83$, and $\mathrm{Pt}_r = 1.48$. To obtain the best estimate of the proper normalization of this 126-neutron peak, we take the ratio of the sum of the observed r-process abundances to the sum of the three calculated r-process abundances. This ratio is $1.53/3.23 = 0.47$. This indicated normalization is sufficiently close to that applying to the lower mass region considered earlier that we finally adopt the normalization parameter for the trans-bismuth atomic weights to lie in the range $0.3 < r < 0.5$.

TABLE 5

COMPARISON OF OBSERVED AND CALCULATED PARTIAL SUMS OF RARE-EARTH ABUNDANCES

Sum	Observed ΣN_r	Calculated ΣN_r	Obs ΣN_r/Calc ΣN_r
$\sum_{n=0}^{6} N_{147+4n}$	0.326	0.979	0.33
$\sum_{n=0}^{7} N_{148+4n}$	0.416	1.174	0.35

From these rather extended considerations of the contributions of the s- and r-processes to primordial lead, we conclude with a tractable expression for the cosmoradiogenic abundances of Pb^{206} and Pb^{207}:

$$\mathrm{Pb}_c^{206}/\mathrm{Pb} = 0.189\,(1 - f_s) - 0.48\, r/\mathrm{Pb}\,,$$

and

$$\mathrm{Pb}_c^{207}/\mathrm{Pb} = 0.206 - (0.095 \pm 0.019)\, f_s - 0.37\, r/\mathrm{Pb}\,. \tag{19}$$

The parameters f_s and r were both concluded to be in the range 0.4 ± 0.1

A look at these two equations shows that the naturally occurring parameters are f_s and r/Pb. Helliwell (1961) has found the solar abundance of lead to be $\mathrm{Pb} = 2.5 \pm 1.0$ on the scale $\mathrm{Si} = 10^6$. This value depends on the correctness of the solar silicon/hydrogen

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ratio of Goldberg, Müller, and Aller (1960), who found Si/H = 3.16×10^{-5}. Employment of the older Si/H ratios, Si/H = 1.95×10^{-5} (Unsöld 1948) and Si/H = 1.32×10^{-5} (Class 1951), would lead to higher values of Pb on the Si = 10^6 scale. Using Helliwell's value for the lead abundance and the value $r = 0.4 \pm 0.1$, we calculate that the parameter r/Pb has the value 0.16 ± 0.07. This value of r/Pb would conceivably be considerably smaller if the older Si/H ratios are actually more nearly correct than that of Goldberg *et al.* We will give some weight to the older Si/H ratios by assuming that Pb = 2.5 is a lower limit on the lead abundance, and its most probable value as well. Although this is a highly doubtful procedure from a purely statistical point of view, we believe it to be a physically reasonable one. It is appropriate to emphasize at this point that the method of this paper depends upon a well-determined solar lead abundance, and it is hoped that further research into the solar spectrum and the theory of line formation will point the way to an unequivocal value for Pb. With our assumption about Pb, the parameter r/Pb is less than 0.20, has a most probable value of 0.16, and may be considerably smaller.

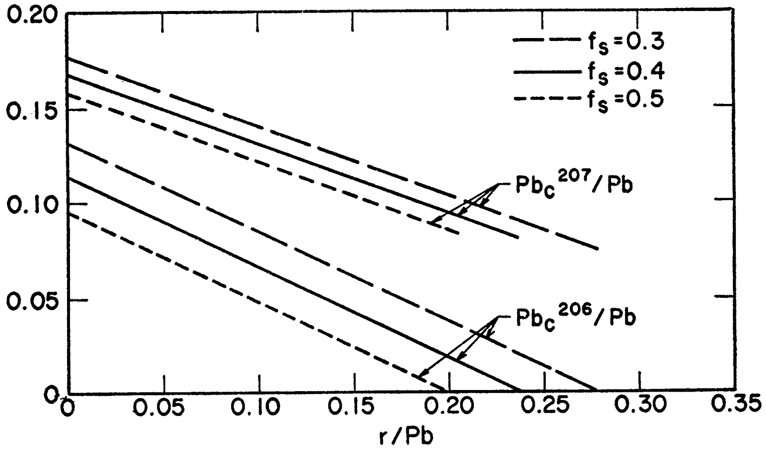

FIG. 7.—The fractions Pb_c^{207}/Pb and Pb_c^{206}/Pb for various values of the two parameters f_s and r/Pb. Tentative estimates indicate that $f_s = 0.4 \pm 0.1$ and $r/Pb = 0.16 \pm 0.04$.

The values of Pb_c^{206}/Pb and Pb_c^{207}/Pb as a function of r/Pb are shown in Figure 7 for three values of f_s = 0.3, 0.4, and 0.5. Since Pb_c^{206}/Pb is a positive number, we see from this figure another indication that r/Pb cannot much exceed 0.20 for these likely values of f_s.

d) The Abundance of Uranium

We now turn our attention to the chronological relationships derived earlier and ask how they may best be employed to obtain useful information. The first of these will be used in the following form:

$$R = \frac{Pb_c^{207}/Pb + U^{235}/Pb}{Pb_c^{206}/Pb + U^{238}/Pb} = \frac{Pb_c^{207}/Pb + 0.236\ U/Pb}{Pb_c^{206}/Pb + 0.764\ U/Pb}. \qquad (20)$$

The primordial U/Pb abundance ratio is not well enough known to calculate the production ratio R from this formula, and indeed there is no assurance that it will ever be well known experimentally. For this reason, this equation will be more useful as a means of computing U/Pb from an assumed range of values for R. In fact, we will assume for the remainder of this paper that $1.4 \leq R \leq 1.8$. This is a rather broad range of R and is chosen to include the calculated production ratios $R = 1.65$ (Fowler and Hoyle 1960) and $R = 1.45$ (Cameron 1962). What we propose to show is that even such a broad range of assumed values of R can lead to sizable restrictions on both the U/Pb ratio and the chronological model of galactic nucleosynthesis.

We first note that, for each model of exponential nucleosynthesis, the assumed range of R delimits a range of times for the beginning of nucleosynthesis (see Fig. 4). For the same models and times, Figure 5 shows the calculated values of $R\ Pb_c^{206}/Pb_c^{207}$. These two relationships may be employed in the following way: equation (20) for R may be rearranged to read

$$\frac{U}{Pb}=\left(\frac{1-R\ Pb_c^{206}/Pb_c^{207}}{0.764R-0.236}\right)\frac{Pb_c^{207}}{Pb}. \qquad (21)$$

Now consider for a moment any exponential model of nucleosynthesis; that is, one in which the rate of r-process nucleosynthesis at the time of solar-system formation is some specified fraction of the initial rate in the Galaxy. Then for each value of R, the time of the commencement of nucleosynthesis is specified by Figure 4, and for that value of time the quantity $R\ Pb_c^{206}/Pb_c^{207}$ is specified from Figure 5. From the equation above, it is clear that the relationship between U/Pb and Pb_c^{207}/Pb is thereby completely determined *for that model and that value of R*. This procedure may be performed for each model and for each value of R in the expected range of R. We conclude, therefore, that U/Pb may be computed for each exponential model if Pb_c^{207}/Pb is known.

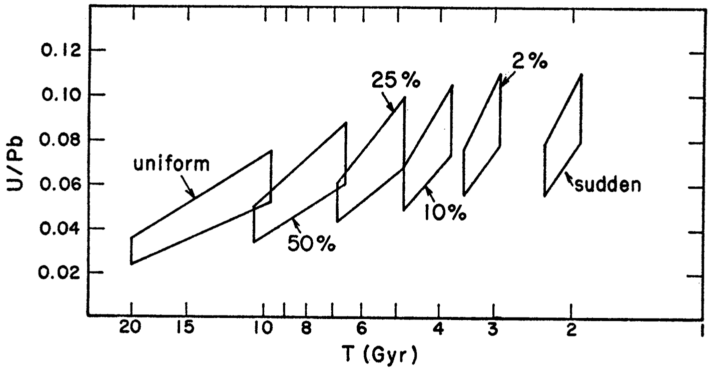

FIG. 8.—The U/Pb abundance ratio for the exponential models of galactic nucleosynthesis versus the time of the being of nucleosynthesis. For each exponential model, the consistent values of U/Pb and T must lie interior to the trapezoid for that model.

Of course, Pb_c^{207}/Pb is not exactly known, but an inspection of Figure 7 reveals that, for the likely values of r/Pb and f_s, $Pb_c^{207}/Pb = 0.12 \pm 0.02$. This simple estimate of Pb_c^{207}/Pb makes possible a two-dimensional representation wherein, for each exponential model, the possible range of U/Pb may be plotted against the possible range in time of the beginning of nucleosynthesis. Figure 8 shows the results of such a calculation. For each model, the possible values of U/Pb and T lie interior to the trapezoid for that model. It is obvious that for all models, and for any R in the range 1.4–1.8, the ratio U/Pb can differ by no more than a factor of two from the value U/Pb = 0.052. This latter value is quite close to the primordial value U/Pb = 0.050 (Clayton 1963) expected from the concordant chronology of nucleosynthesis given by Fowler and Hoyle (1960). It is apparent that the smallest values of U/Pb are consistent with a relative uniform rate of nucleosynthesis over periods of about 10^{10} years before solar-system formation, whereas the highest U/Pb values indicate rates of nucleosynthesis more strongly peaked in the initial epoch, which is correspondingly more recent. The U/Pb ratio is a poor indicator of the chronology of nucleosynthesis because it is relatively insensitive to chronologies. This lack of sensitivity, on the other hand, allows the confident prediction for exponential chronologies that $0.026 < U/Pb < 0.104$. Only if R is less than 1.4 or greater than 1.8 can this range of U/Pb values be violated. To say that it is *probable* that U/Pb falls outside the indicated range (a much stronger statement) would require $R < 1.0$ or $R > 2.0$.

Finally, in order that no confusion over this point arise, we note that Clayton (1963) quoted a much smaller error, $U/Pb = 0.0499 \pm 0.0044$. That calculation was based upon a specific model of the chronology of nucleosynthesis, and the error quoted there does not include any error in the model used, which was accepted as being the best available. The present investigation reveals how much U/Pb may vary with extreme changes in the model. This calculation of the U/Pb abundance ratio, moreover, in contrast to other calculations of that ratio, does not depend explicitly upon the absolute production rate of uranium in supernovae.

Some amplification of this distinction will be illuminating. Fowler and Hoyle (1960) calculated the primordial abundance of uranium on the scale $Si = 10^6$ to be $U = 0.293$. This calculation employed their model of the chronology of nucleosynthesis and the absolute r-process yield of uranium when normalized in the same manner as the r-process calculation of Burbidge *et al.* (1957). We have seen that this normalization is too large by a factor of about 2.5. Making this reduction would indicate that $U = 0.117$. In a more recent reinvestigation by this technique, Hoyle and Fowler (1963*b*) conclude $U = 0.088$. Clayton (1963) used data on the neutron-capture cross-sections of the lead isotopes and the chronology of Fowler and Hoyle (1960) to conclude that the appropriate normalization parameter for the r-process production of uranium is related to the abundance of lead by the equation $r' = 0.17\ Pb = 0.17 \times 2.5 = 0.42$. Multiplying Fowler and Hoyle's production rate by this normalization yields $U = 0.123$. The present work has used neither Fowler and Hoyle's production rates for uranium nor their chronology of nucleosynthesis. The primordial abundance of uranium inferred from the enrichment of the lead isotopes is within a factor of two of $U = 0.052\ Pb = 0.052 \times 2.5 = 0.130$. That these different viewpoints converge to roughly the same value of U lends strength to the correctness of that number. Since U (now) = 0.38 U, we conclude that the present solar abundance of uranium is within a factor of 2 of the value U(now) = 0.048 on the scale $Si = 10^6$.

It must be recognized that a present value of $U = 0.048$ is about six times larger than the value inferred from the average uranium and thorium concentrations in chondrites (Bate, Huizenga, and Potratz 1959; Goles and Anders 1962). Either the chondrites have fractionated U relative to Si, or our calculation of U involves an incorrect assumption. In the latter regard, we would like to return to the idea that the average galactic nucleosynthesis may have been augmented in solar-system material by a discrete event shortly before the formation of the solar system. *If* such an event occurred, it would have considerably increased the U^{235}/U^{238} ratio without adding anything to the cosmoradiogenic lead abundance. Under those conditions, a much larger fraction of the galactic uranium may have decayed and lower U/Pb ratios may be obtained. The clue to this possibility lies in whether the uranium isotope chronology alone is concordant with the cosmoradiogenic chronologies presented in this paper for simple exponential models of nucleosynthesis. We feel that a quantitative treatment of this idea is inappropriate at the present time, however, due to the lack of sufficient data on the neutron-capture cross-sections stressed in this work.

e) The Pb_c^{206}/Pb_c^{207} *Ratio as a Chronometer*

From the equations of earlier sections, we have seen that, for any assumed exponential model, a value of R determines both the time T and Pb_c^{206}/Pb_c^{207}. Figure 9 shows this two-dimensional relationship between Pb_c^{206}/Pb_c^{207} and T for each model. If we then restrict R to the range $1.4 < R < 1.8$, only a corresponding segment of each model is allowable. The envelopes of those segments for all models are shown in dashed lines in

Figure 9, the upper envelope being for $R = 1.4$ and the lower for $R = 1.8$. The shaded region in between then represents all the possible values in the Pb_c^{206}/Pb_c^{207}, T-plane that are consistent with $1.4 < R < 1.8$. The rather sizable increase of Pb_c^{206}/Pb_c^{207} with age is the essential clue to obtaining a chronometer from the composition of primordial lead. The possibility of exploiting this correlation will clearly depend on our ability to compute the Pb_c^{206}/Pb_c^{207} ratio.

From the expressions for Pb_c^{206}/Pb and Pb_c^{207}/Pb derived as equation (19), it follows that

$$\frac{Pb_c^{206}}{Pb_c^{207}} = \frac{0.189(1 - f_s) - 0.48 r/Pb}{0.206 - (0.095 \pm 0.019) f_s - 0.37 r/Pb}. \qquad (22)$$

We have plotted these values of Pb_c^{206}/Pb_c^{207} in Figure 10 for three values of f_s and for $0.10 < r/Pb < 0.20$. It is evident that almost any value of Pb_c^{206}/Pb_c^{207} is obtainable, depending upon the values of f_s and r/Pb; moreover, Pb_c^{206}/Pb_c^{207} bears an approximately linear inverse relationship to both f_s and r/Pb, having its maximum value for small f_s and small r/Pb. Figure 10 points out clearly the need for secure estimates of f_s and r/Pb. From an experimental point of view, these values depend most critically on the values of Pb^{208} neutron-capture cross-section and the abundance of lead. We showed earlier that $f_s \simeq 0.5\ \bar{\sigma}(208)$ is a good approximation to the value of f_s, but $\bar{\sigma}(208)$ has not been accurately measured. Since $r \sim 0.4$, $r/Pb \sim 0.16$ if we use the value $Pb = 2.5$ relative to $Si = 10^6$. Continued efforts to carefully establish the solar Pb/H and Si/H ratios are advocated in order to minimize uncertainties in the value of r/Pb.

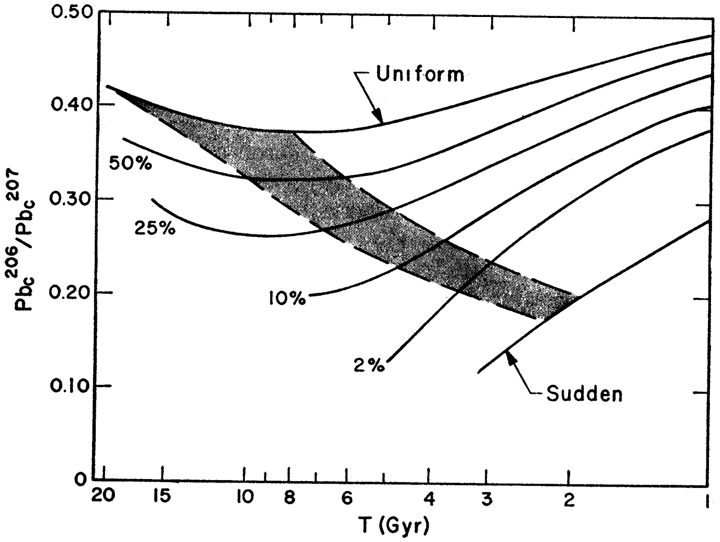

FIG. 9.—The ratio Pb_c^{206}/Pb_c^{207} as a function of the time of commencement of the exponential models of galactic nucleosynthesis. Restricting the uranium production ratio to $1.4 < R < 1.8$ limits the commencement time for each model by the relationship shown in Fig. 4. The shaded region between the dashed-curve envelopes shows the allowable region for this restriction on R; the upper envelope is for $R = 1.4$, the lower one for $R = 1.8$. The resulting increase of Pb_c^{206}/Pb_c^{207} with age provides the basis for a chronometer.

As an example of how the analysis may go, we adopt the most probable estimates of this paper, $f_s = 0.4$, $r/Pb = 0.16$. The indications of Figure 10 are then that $Pb_c^{206}/Pb_c^{207} \simeq 0.33$. This value of Pb_c^{206}/Pb_c^{207} in Figure 9 implies times for the beginning of nucleosynthesis ranging from 6 to 10 billion years before the formation of the solar system as well as exponential models for which the rate of nucleosynthesis at solar-system formation ranges from 30 to 50 per cent of its initial value. It will be clear that this is only a most probable estimate at the present time, but this method itself must not be overlooked in the total of evidence regarding the history of galactic nucleosynthesis. As a final point here, we note that this tentative solution is not in disagreement with the chronology of Fowler and Hoyle, who found $T \sim 7 \times 10^9$ years for a 50 per cent exponential model.

f) The Abundance of Bismuth

We wish to digress again at this time to briefly consider one problem associated with the foregoing analysis. The Bi^{209} abundance has resulted from the same mechanisms discussed earlier in connection with the Pb^{206}, Pb^{207}, and Pb^{208} abundances. We may again write

$$Bi^{209} = Bi_s^{209} + Bi_r^{209} + Bi_c^{209} . \tag{23}$$

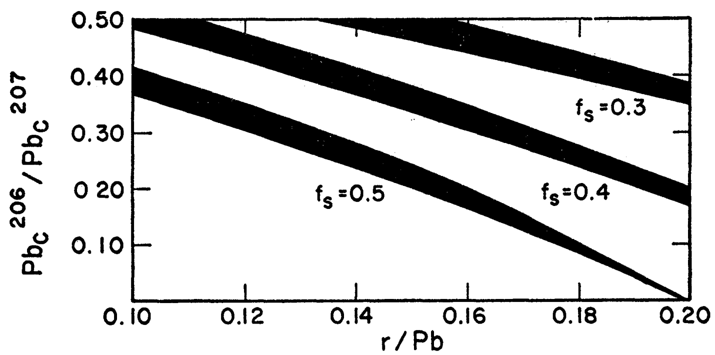

FIG. 10.—The ratio Pb_c^{206}/Pb_c^{207} for likely values of the parameters f_s and r/Pb. A tentative estimate of the most probable values, $f_s = 0.4$ and $r/\mathrm{Pb} = 0.16$, yields $Pb_c^{206}/Pb_c^{207} = 0.33$. The width of the shaded band for each value of f_s reflects the uncertainty in the denominator of eq. (22), which in turn resulted from $Pb_s^{207}/Pb_s^{206} = 0.5 \pm 0.1$ in eq. (15).

Calculations of these three terms are as follows:

1. From s-process equilibrium, $\bar{\sigma}(209)\, Bi_s^{209} = \bar{\sigma}(208)\, Pb_s^{208}$. Since something like 80 per cent of Pb^{208} must be due to the s-process, we can estimate $Pb_s^{208} = (0.47 \pm 0.05)$ Pb. Then $Bi_s^{209}/\mathrm{Pb} = (0.47 \pm 0.05)\, \bar{\sigma}(208)/\bar{\sigma}(209)$.

2. From the analysis of the short-lived progenitors of Bi^{209} created in the r-process, it follows that $Bi_r^{209}/\mathrm{Pb} = 0.32\, r/\mathrm{Pb}$.

3. From the analysis of Fowler and Hoyle (1960) we see that U^{235} is synthesized in the r-process in abundance greater than that of Np^{237} by the factor $0.662/0.622 = 1.06$. For reasonable models of nucleosynthesis beginning around 6 billion years before solar system formation, we know that something like 90 per cent of the original U^{235} must have decayed to Pb^{207} by the time of the solar-system formation. Since all the Np^{237} will have decayed to Bi^{209} in the same interval, we may conclude that $Bi_c^{209} = (0.95 \pm 0.05)$ Pb_c^{207}. We showed earlier that $Pb_c^{207}/\mathrm{Pb} = 0.12 \pm 0.02$, so $Bi_c^{209}/\mathrm{Pb} = 0.11 \pm 0.02$.

Summing these contributions gives

$$\mathrm{Bi/Pb} = (0.47 \pm 0.05)\, \bar{\sigma}(208)/\bar{\sigma}(209) + 0.32\, r/\mathrm{Pb} + 0.11 \pm 0.02 . \tag{24}$$

Evaluation of this expression is again stalled by uncertainties in the cross-section ratio. However, on the anticipation that $\bar{\sigma}(208)/\bar{\sigma}(209) = 0.5 \pm 0.1$, and using again $r/\mathrm{Pb} = 0.4 \pm 0.1$, we may tentatively estimate that

$$\mathrm{Bi/Pb} \simeq 0.5 \pm 0.1 .$$

This very high abundance of bismuth serves as a dramatic indication of the extent to which bismuth has been lost in the formation of solid bodies.

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g) The Steady-State Model of Solar Abundances

It has been proposed as a possible alternative to autonomous galactic synthesis that the solar system may have formed from well-mixed intergalactic gas that was absorbed by the Galaxy about 5 × 10^9 years ago (Fowler and Hoyle 1960).

Both the Re^{187}-decay and the U-Pb relationship may be tested for concordance with such a model. The self-consistent solution would demand a relatively high cross-section ratio for the osmium isotopes, $\sigma(186)/\sigma(187) = 0.6 \pm 0.1$, and a high primordial uranium abundance, $U/Pb = 0.06 \pm 0.02$.

V. SUMMARY AND CONCLUSIONS

We have introduced two new nuclear methods for dating the time and duration of galactic nucleosynthesis. The first of these involves the beta-decay of Re^{187} to Os^{187}. We have shown that the amount of cosmoradiogenic Os^{187} is determinable. The second method involves both the uranium isotope ratio and the cosmoradiogenic abundances of Pb^{206} and Pb^{207}. We have shown that these cosmoradiogenic abundances may ultimately be computed with sufficient accuracy to allow a chronological estimate. It is unfortunate that neither of these methods yields definitive results at the present time. The uncertainty is largely due to insufficient knowledge of some measurable properties of certain nuclei, although poor knowledge of key abundances also contributes. Because of the importance of continued research to eliminate these insufficiencies, we enumerate here some of the major research problems applicable to the over-all line of attack outlined in this paper.

1. Average neutron-capture cross-sections for a Maxwell-Boltzmann neutron velocity spectrum corresponding to $kT = 30$ to 60 keV are sorely needed for these key nuclei, in decreasing order of importance: Os^{186}, Os^{187}, Pb^{208}, Pb^{204}, Pt^{192}, and Bi^{209}.
2. A thorough investigation of the Re^{187} decay is needed to determine its half-life, end-point energy, and the possible importance of bound-state decay in its total decay rate.
3. The Os/Re abundance ratio is needed to determine the amount of radiogenic Os^{187}. To determine the amount of radiogenic lead, the abundances Pb, Te, Os, Ir, and Pt on the $Si = 10^6$ scale are required.
4. More accurate meteoritic isochrones for the Re^{187}-Os^{187} decay are needed to secure the primordial Os^{187}/Os^{186} ratio.

In the absence of secure determinations of these important quantities, we have adopted "most likely values" for them—values inferred from the systematics of the body of existing facts. We have tentatively estimated the chronological implications of these values. The Re^{187}-Os^{187} decay favors a time for the beginning of galactic nucleosynthesis in the range of 6–14 billion years before the formation of the solar system. The U-Pb-decays favor a time in the range 6–10 billion years. By comparison, the concordant U-Th chronology of Fowler and Hoyle (1960) determined a most likely time of 7 billion years. Although the commencement time is still not accurately determined, the evidence is now compelling that nucleosynthesis must have begun at least 5 × 10^9 years before the formation of the solar system. Models of nucleosynthesis shortly before solar-system formation simply do not allow enough time for the build-up of daughter-product abundances.

We examined also the idea that the solar system may have formed from steady-state abundances injected into the Galaxy less than 1 billion years before the time of solar formation. It was found that neither method contradicted this idea at our present stage of knowledge; however, the possibility remains that the Re^{187}-Os^{187} decay may ultimately do so when the Os^{186} and Os^{187} cross-sections are measured, and the U-Pb method may ultimately do so if strong geochemical arguments for low U/Pb values can be presented.

It is to be hoped that the experimental reduction of the number of unknown or poorly known quantities will allow these two methods to join the older U-Th method as nuclear clocks for the chronology of nucleosynthesis. Ultimately, a concordant solution of all these techniques should make the correct chronology secure. In fact, the demand for concordance may even illuminate structure in the history of nucleosynthesis.

I am indebted to Dr. R. L. Macklin for permission to use his measurements of neutron-capture cross-sections in advance of publication, and to Professor William A. Fowler for many discussions of *r*-process production ratios. For helpful comments I am grateful to Professor J. L. Greenstein, Dr. G. G. Goles, Dr. W. Nichiporuk, and Dr. B. Hirt.

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