

Neutron-Capture Cross Sections for Osmium Isotopes and the Age of the Universe*

The neutron-capture cross sections for the light osmium isotopes ^{186}Os and ^{187}Os recently have been measured in the energy range up to 150 keV. The knowledge of these cross sections enables us to calibrate the $^{187}\text{Re} \rightarrow ^{187}\text{Os}$ nuclear β -decay clock and thus to make a new radiogenic determination of the age of the Universe. The value thus determined, with certain assumptions, is 17 ± 3 billion years. This value, larger than had been believed heretofore, is in accord with the age of recently discovered galaxies; however, it exceeds the most recent determination of the Hubble time, with important cosmological implications.

INTRODUCTION

According to the currently accepted view,¹ the Universe began with a "Big Bang," a singularity in time and density (or perhaps temperature). Some 1 to 2 b.y. (billion years) later,² the first galaxies, including our own Milky Way, were formed out of the debris of the Big Bang. It is well known that our solar system condensed out of interstellar material (the ejecta from previous supernovae in our galaxy) about 4.7 b.y. ago. Thus, a measure of the time interval between the formation of our galaxy and the condensation of our solar system is needed to know the age of the Universe A_U . This time interval Δ , the difference between the age of our galaxy A_G and the age of the solar system A_S , is the duration of nucleosynthesis, the time interval during which the matter of which the solar system is composed was being processed in stars; thus A_G is also the age of the elements. Figure 1 shows these time

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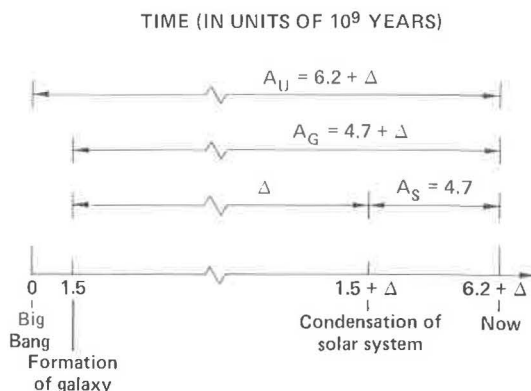


FIGURE 1 The time intervals involved in cosmology, showing the critical importance of a knowledge of the value of the duration of nucleosynthesis Δ for the determination of the age of the Universe A_U . (A_S is the age of our solar system and A_G is the age of our galaxy.).

intervals. This report describes a recent experimental effort to determine Δ accurately, and through it, A_U .

The basic mechanisms for the formation of the elements have been delineated in the classic paper of Burbidge *et al.*³ Figure 2, from Ref. 3, illustrates these mechanisms on a chart of the nuclides. The elements heavier than iron (where the binding energy per nucleon reaches its maximum) are formed by successive neutron capture, which can proceed on either a slow time scale (the s process) or a rapid one (the r process). The source of neutrons has not been determined unambiguously, but it is thought that the $^{22}\text{Ne} + ^4\text{He} \rightarrow ^{25}\text{Mg} + \text{n}$ reaction plays the most important role.⁴ The s process proceeds along the line of β stability, terminating at bismuth, and takes place primarily in helium-burning stars; the r process departs substantially from the locus of stable nuclei, is exclusively responsible for production of the elements heavier than bismuth, and most likely takes place primarily in supernovae.

The most important nuclear dating method that is based upon the r process is the U-Th technique of Fowler and Hoyle.⁵ This method consists of calculating the production ratios for ^{232}Th , ^{235}U , and ^{238}U in supernova events, and then by measuring the current abundance ratios of the same nuclei, arriving at a chronology of supernova events. Thus, with the use of a model of the time dependence of such events (the same exponential model as is used here), one obtains the duration of

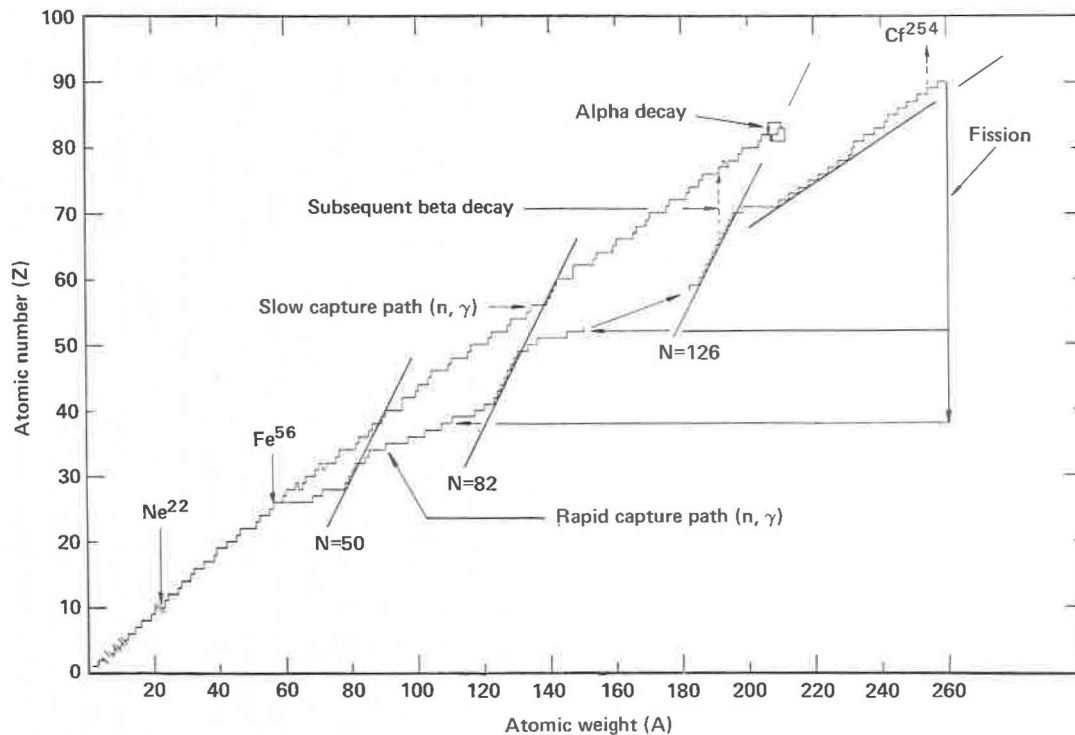


FIGURE 2 Representation, on a chart of the nuclides, of the slow (s) and rapid (r) processes for the formation of the elements heavier than iron (from Ref. 3).

nucleosynthesis and hence the age of the galaxy. There are two important uncertainties connected with this method (other than the model for supernova frequency employed), namely (1) that the production ratios of the chronometer nuclei cannot be measured directly and therefore must be calculated, and this calculation depends upon certain input parameters that are very hard to test and (2) that the half-lives of the chronometer nuclei are comparable to (and not much larger than) their age.

THE $^{187}\text{Re} \rightarrow ^{187}\text{Os}$ CLOCK

In 1964, Clayton⁶ suggested the use of a process which does not depend critically upon a knowledge of the r-process production rates. Rather it depends mainly upon the s process, which rests upon a more firm theoretical footing. Clayton's suggestion involves the β -decay of ^{187}Re (which has a half-life of ~ 43 billion years⁷) to ^{187}Os . Because both ^{186}Os and ^{187}Os are pure s-process nuclei (i.e., they are shielded by ^{186}W and ^{187}Re , respectively, from the r process) and because these heavy metals are present in a relatively undisturbed state in primordial meteorites, one has a favorable situation. It was Clayton's assertion that a nuclear chronometer based upon the s process in general, which would not depend upon the assumption (1) above, would be more reliable; and that the $^{187}\text{Re} \rightarrow ^{187}\text{Os}$ chronometer, in particular, because of the very long half-life of ^{187}Re , would be best.

An s-process chronometer, on the other hand, depends upon the "local" assumption, namely, that the product $N\bar{\sigma}$ is constant for adjacent s-process nuclei, where N is the s-process abundance for a given nuclear species and $\bar{\sigma}$ is its Maxwellian-averaged neutron-capture cross section at the temperature appropriate to the stellar site of the s process ($kT \approx 30$ keV). However, this assumption has been well justified by the work of Macklin *et al.*⁸ for the strontium, zirconium, tin, and samarium isotopes; it probably holds even more rigorously for the osmium isotopes, which are far from any nuclear shell closure. Hence, little uncertainty should result from the use of the local assumption.

Figure 3 shows the portion of the chart of the nuclides in the vicinity of the osmium and rhenium isotopes. Both the s-process and r-process paths are shown. It can be seen that ^{187}Re is an r-process nucleus and that both ^{186}Os and ^{187}Os are shielded from the r process, so that the

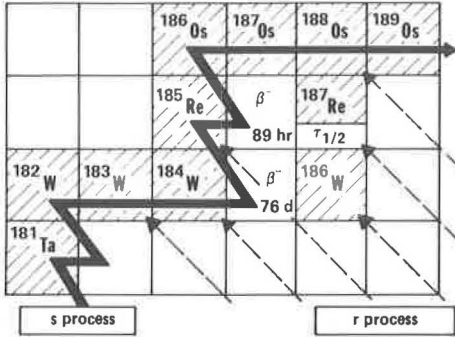


FIGURE 3 The s-process (heavy line) and r-process (dashed lines) paths in the vicinity of the osmium isotopes (from Ref. 6). The shaded boxes represent stable nuclei except for ^{187}Re ($\tau_{1/2} \approx 43$ b.y.). It can be seen that ^{186}Os and ^{187}Os are shielded from the r process by ^{186}W and ^{187}Re , respectively.

synthesis of ^{186}Os and ^{187}Os initially involves only the s process. If we denote the s-process abundances of ^{186}Os and ^{187}Os as N_s^{186} and N_s^{187} and the radiogenic component of ^{187}Os resulting from the β decay of ^{187}Re as N_{rad}^{187} , then the total abundances can be written as

$$N^{186} = N_s^{186} \text{ and } N^{187} = N_s^{187} + N_{\text{rad}}^{187}. \quad (1)$$

Using the local assumption $N_s^{186} \bar{\sigma}_{186} \cong N_s^{187} \bar{\sigma}_{187}$ and Eqs. (1), we can express the ratio of N_{rad}^{187} to its parent ^{187}Re abundance N_{Re}^{187} as

$$R = \frac{N_{\text{rad}}^{187}}{N_{\text{Re}}^{187}} = \frac{(N^{187}/N_{\text{Os}}) - f(\bar{\sigma}_{186}/\bar{\sigma}_{187})_{\text{lab}}(N^{186}/N_{\text{Os}})}{N_{\text{Re}}^{187}/N_{\text{Re}}} \frac{N_{\text{Os}}}{N_{\text{Re}}} \quad (2)$$

where N_{Os} and N_{Re} are the elemental abundances of osmium and rhenium. The measured Maxwellian-averaged laboratory cross-section ratio $(\bar{\sigma}_{186}/\bar{\sigma}_{187})_{\text{lab}}$ needs to be multiplied by a correction factor f in order to account for the fact that the osmium nuclei in a stellar environment exist in excited states as well as in their ground state. In particular, the 9.8-keV excited state of ^{187}Os plays an important role here.

We now assume (as did Clayton⁶) that r-process nucleosynthesis (which formed ^{187}Re) began at a time Δ before the condensation of the solar system and decreased exponentially at a rate λ_A . Therefore λ_A is a measure of the supernova rate in the galaxy. If λ_B denotes the β -decay rate of ^{187}Re , one can express the ratio $N_{\text{rad}}^{187}/N_{\text{Re}}^{187}$ at the time of solar-

system condensation, using the Bateman equations for radioactive growth and decay, as

$$R = \frac{N_{\text{rad}}^{187}}{N_{\text{Re}}^{187}} = \left[\frac{\lambda_A - \lambda_B}{\lambda_A} \exp(\lambda_B \Delta) \frac{1 - \exp(-\lambda_A \Delta)}{1 - \exp[-(\lambda_A - \lambda_B) \Delta]} \right] - 1. \quad (3)$$

Two extreme cases of this model are (1) sudden synthesis (a single supernova event), for which $\lambda_A \rightarrow \infty$ and Eq. (3) becomes $N_{\text{rad}}^{187}/N_{\text{Re}}^{187} \simeq \lambda_B \Delta$, and (2) uniform synthesis (a constant rate of supernova events), for which $\lambda_A \rightarrow 0$ and Eq. (3) becomes $N_{\text{rad}}^{187}/N_{\text{Re}}^{187} \simeq \lambda_B \Delta/2$. Fowler⁹ suggested a kind of "happy medium" value for λ_A , based upon the idea that the supernova rate in our galaxy was higher when it was younger, corresponding to the existence in earlier eons of a larger number of massive stars (which are more likely to end their existence as supernovae, and do so on a shorter time scale, than less massive stars). This value, $\lambda_A^{-1} = 0.43 \Delta$, which is used here, has been used as well for the determination of Δ by the U-Th method.

The isotopic abundance ratios in Eq. (2) can be obtained from meteoritic abundance data,¹⁰ and, when referred back to the time of solar-system condensation using $\tau_{1/2}(^{187}\text{Re}) = 43 \text{ b.y.}$, have the values $N_{\text{Os}}^{187}/N_{\text{Os}} = 0.0125 \pm 0.0006$, $N_{\text{Os}}^{186}/N_{\text{Os}} = 0.0159$, and $N_{\text{Re}}^{187}/N_{\text{Re}} = 0.65$ (the geochemical uncertainties for the latter two quantities are negligible). For the elemental abundance ratio $N_{\text{Os}}/N_{\text{Re}}$, we have chosen to use the value of 13.1 ± 0.6 recommended by Woosley¹¹; this value is obtained from elemental analysis of type-C1 carbonaceous chondrites only, and excludes the (slightly lower) values from other types of meteorites¹⁰ that are thought to be less ancient. The remaining quantities that are critical to the determination of R in Eq. (2) and hence to Δ in Eq. (3) are the cross-section ratio $(\sigma_{186}/\sigma_{187})_{\text{Lab}}$ and its correction factor f . The factor f can be calculated, given certain input data (see below), and in fact this has been done.¹² The cross sections, however, had to be measured.

THE EXPERIMENTS

The key role played by the neutron-capture cross sections for ^{186}Os and ^{187}Os (or rather, by their ratio) in this determination of the age of the Universe also was pointed out by Clayton.⁶ Their measurement, however, would have been beset with formidable experimental difficulties until the sufficiently massive and isotopically pure samples of these rare

osmium isotopes were manufactured specially for the measurements performed by Browne and Berman.¹³ Subsequent measurements using these enriched samples also have been carried out by Browne *et al.*¹⁴ and by Winters *et al.*¹⁵ Thanks to the help of many people and a year-and-one-half effort at the Oak Ridge National Laboratory for the isotopic enrichment of these samples (3.278 g of ¹⁸⁶Os, 78.39% pure, and 2.959 g of ¹⁸⁷Os, 70.96% pure), these measurements were made possible.

The experiment of Refs. 13 was performed at the neutron time-of-flight facility at the Lawrence Livermore National Laboratory Electron-Positron Linear Accelerator. Neutrons were produced in a water-cooled tantalum target struck by a pulsed 115-MeV electron beam from the linac. The energy of the neutrons was determined by their times of flight down an evacuated, collimated flight tube to the samples located ~15 m from the neutron source.

The measurement was performed with samples, cycled appropriately into the neutron beam, of enriched ¹⁸⁶Os, ¹⁸⁷Os, ¹⁸⁸Os, ¹⁸⁹Os, ¹⁹⁰Os, and ¹⁹²Os (since the ¹⁸⁶Os and ¹⁸⁷Os samples were not 100% pure, the other osmium isotopes also had to be measured) enclosed in light-weight beryllium containers, and with an empty container as well. The neutron-capture γ rays were detected by a pair of large deuterated-benzene (C₆D₆) liquid scintillators.

The neutron-capture cross sections for ¹⁸⁶Os and ¹⁸⁷Os measured in this experiment are shown in Figure 4. From these data, the Maxwellian-averaged cross sections for $kT = 30$ keV were obtained; their ratio is $(\bar{\sigma}_{186}/\bar{\sigma}_{187})_{\text{Lab}} = 0.48 \pm 0.04$, and is only slightly dependent upon the choice of temperature near $kT = 30$ keV. The results for this quantity from the subsequent measurements of Refs. 14 and 15 are 0.475 ± 0.075 and 0.478 ± 0.022 , respectively; thus all three results are in excellent agreement with each other. (The measurement of Ref. 14 was done at 25 keV with an iron-filtered neutron beam from the reactor at the National Bureau of Standards; that of Refs. 15 also was a neutron time-of-flight measurement, done at the Oak Ridge Electron Linear Accelerator.)

The correction factor f has been obtained from a Hauser-Feshbach calculation by Woosley and Fowler.¹² This calculation requires as input parameters (among other things) the level spacings and spin distributions in the nuclei which result from neutron capture by ¹⁸⁶Os and ¹⁸⁷Os, namely, ¹⁸⁷Os and ¹⁸⁸Os. The level spacings were obtained from the low-energy data of Ref. 13. The level spin distribution for the (¹⁸⁶Os

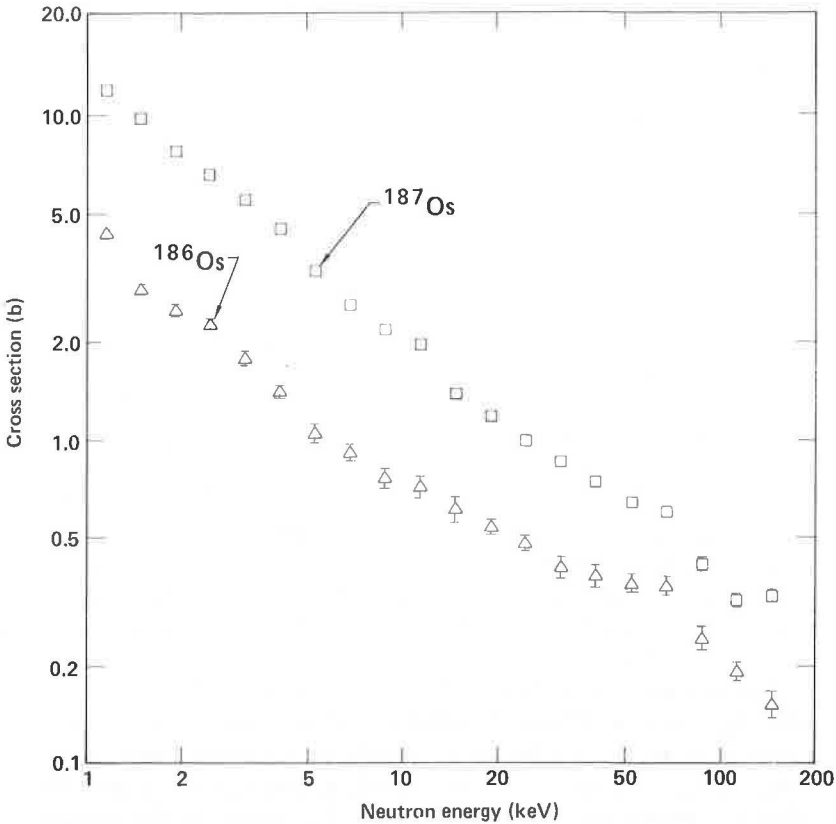


FIGURE 4 The average neutron-capture cross sections versus laboratory neutron energy for ^{186}Os and ^{187}Os (from Ref. 13). The uncertainty for each data point lies within the plotted symbol, except as shown. From these data, the Maxwellian-averaged cross sections for $kT = 30$ keV were obtained; their ratio is the crucial input parameter in Eq. (2).

+ n) system is no problem, since the ground state of the even-even nucleus ^{186}Os has no spin, and at low neutron energies the neutron-capture process is dominated by s-wave capture, so that only spin- $\frac{1}{2}$ levels in ^{187}Os are populated. Because the ground-state spin of ^{187}Os is $\frac{1}{2}$ (so that s-wave neutron capture leads to either spin-0 or spin-1 states in ^{188}Os), however, and moreover because the spin of its important 9.8-keV state is $\frac{3}{2}$ (s-wave capture leads to spin-1 or spin-2 states), it is important to know whether the level-spin distribution in ^{188}Os follows

the usual $(2J + 1)$ rule (J is the spin of the capturing state in the compound system).

To this end, Stolovy *et al.*¹⁶ performed (at Livermore) two-parameter resonance-capture γ -ray measurements on both ^{187}Os and ^{189}Os (which, because its ground-state spin is $\frac{3}{2}$ serves as a mockup for the 9.8-keV first-excited state of ^{187}Os). In this experiment, the resonance neutrons were associated with the γ rays produced by cascade processes to low-lying states in ^{188}Os and ^{190}Os . Since the cascade processes tend to change the spin as little as possible, a γ ray originating from a spin-4 level is more likely than a γ ray originating from a spin-2 level to be associated with a neutron-capture event into a higher-spin state than with one into a lower-spin state. Thus, one can assign spins of the capturing states on the basis of the population of the low-lying states in ^{188}Os and ^{190}Os . The data of Ref. 16 for $^{187}\text{Os} + n \rightarrow ^{188}\text{Os} + \gamma$ are shown in Figure 5. It can be seen from the figure that the population of the spin-0 and spin-1 states follows the $(2J + 1)$ rule; this also turned out to be the case for the spin-1 and spin-2 states in ^{190}Os . In addition, somewhat better values for the average level spacings were obtained from the data of Ref. 16.

THE DETERMINATION OF Δ

With these input data, the calculation of f as a function of temperature¹² results in the curve shown in Figure 6. It can be seen that the value of f at $kT = 30$ keV ($T = 3.6 \times 10^8$ K) is 0.83, and does not depend strongly upon the temperature for values of kT reasonably near 30 keV.

Now we can relate, through Eqs. (2) and (3), the measured value for $(\bar{\sigma}_{186}/\bar{\sigma}_{187})_{\text{Lab}}$ to a value for the duration of nucleosynthesis Δ . The results, corresponding to the three models for the supernova rate in our galaxy discussed above, are shown as the curves in Figure 7. All the experimental data for $(\bar{\sigma}_{186}/\bar{\sigma}_{187})_{\text{Lab}}$ can be combined to give a value of 0.478 ± 0.018 . This is shown as the datum point in the figure plotted on the middle ("happy-medium") curve; it corresponds to a value for Δ of 10.8 ± 1.2 b.y. However, the uncertainty in Δ must be increased because of the uncertainties in f and in $\tau_{1/2}(^{187}\text{Re})$, which have not yet been included. In order to test the value for f , we note that the calculation of Ref. 12 can be extended to predict a value of ~ 1 b for the inelastic neutron-scattering cross section for ^{187}Os to its 9.8-keV state at a neutron

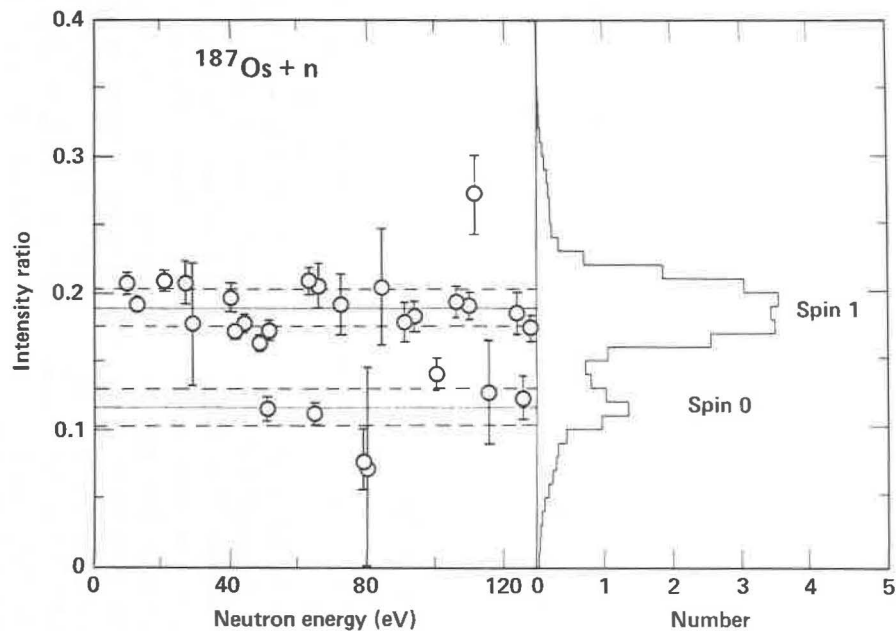


FIGURE 5 The resonance- γ -ray intensity ratio versus resonance energy for ($^{187}\text{Os} + n$), from Ref. 16. The histogram on the right shows the assignment of the resonances into the two spin groups based upon their intensity ratios. This information is needed for the calculation of the correction factor f .

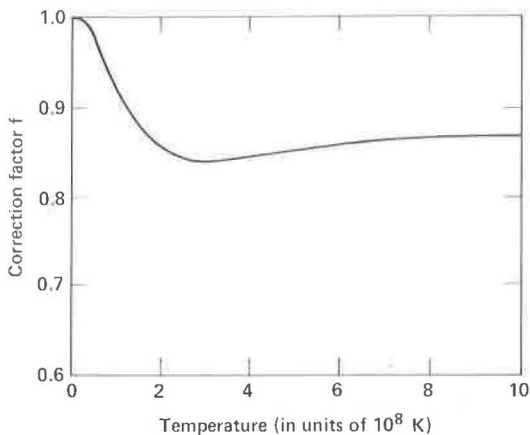


FIGURE 6 The correction factor f which relates the ratio of Maxwellian-averaged cross sections for ^{186}Os and ^{187}Os measured in the laboratory $(\sigma_{186}/\sigma_{187})_{\text{lab}}$ to that which is appropriate for stellar conditions, calculated in Ref. 12. For a temperature near 3.6×10^8 K ($kT \approx 30$ keV), which is appropriate for stellar conditions, the value for f is approximately 0.83.

energy of 30 keV. However, preliminary results from two recent measurements of this $^{187}\text{Os}(n,n')$ cross section disagree with each other: one of these measurements,¹⁷ at 30 keV, sets an upper limit (only) of ~ 0.5 b; the other,¹⁸ at 60 keV, yields a result which would, when extrapolated (which involves some uncertainty), give a value of ~ 1.8 b. The former value would increase f and the latter would decrease f , each by $\sim 20\%$, from the value of 0.83 used here. Thus a better measurement of this cross section at 30 keV is called for.

The half-life of ^{187}Re also is in doubt, partly because its measurement is very difficult and partly because of what appears to be a previous measurement¹⁹ that resulted in a value for $\tau_{1/2}(^{187}\text{Re})$ of 65 ± 13 b.y. (instead of ~ 43 b.y.). Thus, a better measurement of this quantity also is called for; such an effort is underway at LLNL now.²⁰

There are several other potential problems for the $\text{Re} \rightarrow \text{Os}$ method which should be mentioned here. First, in a stellar medium it is possible that the high temperature and density can affect the β -decay rates of the ^{187}Re – ^{187}Os system²¹; the temperature effect is to shorten $\tau_{1/2}(^{187}\text{Re})$ and the density effect is to lengthen it. Next, there is the possibility that the existence of an isomeric state in ^{186}Re (whose spin is 8 and whose half-life is 2×10^5 yr)²² can “short-circuit” the s process there

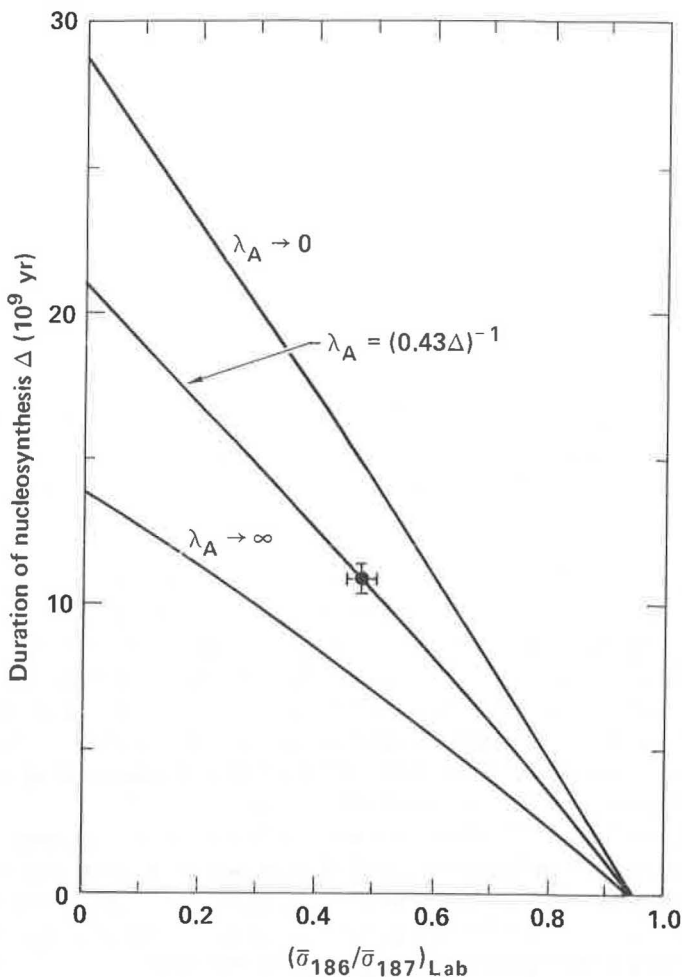


FIGURE 7 The duration of nucleosynthesis Δ prior to solar system condensation plotted as a function of the Maxwellian-averaged laboratory capture-cross-section ratio for ^{186}Os and ^{187}Os . The three models discussed in the text for the supernova rate λ_A are: uniform ($\lambda_A \rightarrow 0$); exponential [$\lambda_A = (0.43\Delta)^{-1}$ —Ref. 9]; and sudden ($\lambda_A \rightarrow \infty$). The datum point represents the combined results of the measurements of Refs. 13–15. The attached error limits include the experimental uncertainties, but not those in $\tau_{1/2}(^{187}\text{Re})$ or in f (see text).

and result in the production of some s-process ^{187}Re (of course, the high spin of the isomeric state argues against this possibility). Finally, there is some controversy regarding the value of the elemental abundance ratio $N_{\text{Os}}/N_{\text{Re}}$ (for instance, the value 13.53 has been given recently²³). Further discussion of these and other related questions can be found in Refs. 24, and they do indeed merit further investigation; meanwhile, although it may not appear that the above analysis will be seriously jeopardized by any of these considerations, one always should be wary of possible surprises.

Factoring "reasonable" uncertainties for f and $\tau_{1/2}(^{187}\text{Re})$ into the uncertainty for Δ , we get a "best" result (at the present time) of $\Delta = 10.8 \pm 2.2$ b.y. This result is almost concordant with the value of 6.1 ± 2.3 b.y. obtained with the U-Th method,⁵ but this is so mainly because of the large uncertainty associated with the determination of f . This "best" value for Δ is also in good agreement with the value of 9.5 b.y. (but with large uncertainties) for the duration of s-process nucleosynthesis recently obtained by Beer and Käppeler²⁵ using the purely s-process chronometer ^{176}Lu .

THE AGE OF THE UNIVERSE

When we add Δ to A_s we get $10.8 + 4.7 = 15.5 \pm 2.3$ b.y. for the age of the galaxy A_G . This is concordant (but not comfortably) with the value $A_G = 11 \pm 3$ b.y. obtained by Iben²⁶ from the globular-cluster method (which attempts to date globular clusters, which are perhaps the oldest objects in our galaxy, but which has serious associated uncertainties⁹). Far more important, Spinrad *et al.*²⁷ recently have reported spectral and photometric observations of two faint galaxies whose ages they estimate to be between 15 and 18 b.y. Finally, when we add 1.5 ± 0.5 b.y. to A_G to account for the time for galaxy formation after the Big Bang² we get $A_U = 17 \pm 2.5$ b.y.; but this uncertainty probably should be increased to ± 3 b.y. in order to encompass the additional uncertainties noted above that cannot be evaluated explicitly at the present time.

The Universe is expanding, as ascertained by observations of the proportional relationship between the distance from us of an astronomical object and its red shift: the farther an object is from us, the farther its radiation spectrum is shifted to longer wavelengths. The propor-

tionality constant is called the Hubble constant, and its inverse the Hubble time. The Hubble time T_H would be equal to A_U if the Universe has been expanding at a constant rate since its origin; that is, T_H represents an upper limit for A_U if the Universe is not decelerating at all. The most recent “conventional” value for T_H is 16.6 ± 1.7 b.y.²⁸ This value for T_H , which is nearly the same as our value for A_U , implies little or no deceleration, which implies in turn that the Universe is “open,” that is, it will continue to expand forever. This behavior is represented by the dashed line in Figure 8, which is a schematic plot of the scale of the Universe as a function of time. If the value for T_H were to have been much larger than A_U , this would have implied a “closed” universe, that is, one which would eventually contract upon itself and at some future time end its existence in an event analogous to the inverse of the Big Bang, namely, a “Big Crunch.” This behavior is represented by

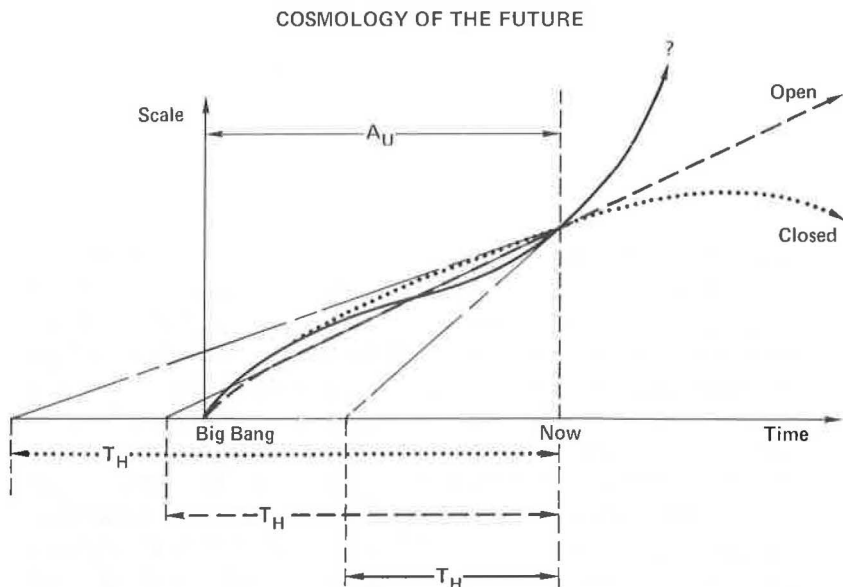


FIGURE 8 Scenarios for the evolution of the scale of the Universe with time for various relative values of the Hubble time T_H and the age of the Universe A_U (assuming the validity of the Big-Bang theory of the origin of the Universe). The dashed line (for $T_H \approx A_U$) represents an open universe, which expands (at a gradually decreasing rate) forever; the dotted line (for $T_H \gg A_U$) represents a closed universe, which (eventually) contracts upon itself; the solid line (for $T_H < A_U$) represents the possible evolution of a universe in which Einstein's cosmological constant is greater than zero.

the dotted line in Figure 8. Thus, the near equality of T_H and A_U means that the Universe will end "not with a bang, but a whimper."

All of this is based upon a universe in which our "ordinary" ideas about gravitation hold, corresponding to a null value for the cosmological constant in Einstein's equations of general relativity. However, there is recent evidence²⁹ that the Hubble time might be substantially smaller than the value given by Sandage and Tammann.²⁸ If this new value for T_H (≈ 10 b.y.) were to be substantiated, then the cosmological implications of a value for T_H that is (considerably) *smaller* than the value of $A_U \approx 17$ b.y. deduced here would be of great importance indeed. The evolution of the Universe then would be represented schematically by the solid line in Figure 8; this line shows a mutual *repulsion* of the objects in the Universe at sufficiently large distances, corresponding to a positive value for Einstein's cosmological constant. This is as if there were a cosmic undertow, strong enough to eventually overcome gravitational attraction, sucking us out into space. The only alternative to this behavior (one to which few astrophysicists would subscribe at present) is that the hot Big-Bang theory is itself no longer valid as a description of the past history of the Universe.

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