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# Ultra-Heavy Cosmic Rays: Theoretical Implications of Recent Observations

J. B. BLAKE  
Space Sciences Laboratory  
The Aerospace Corporation

K. L. HAINESACH  
Lawrence Livermore Laboratory

and

D. N. SCHRAMM and J. D. ANGLIN  
Enrico Fermi Institute  
University of Chicago

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IMPLICATIONS OF RECENT OBSERVATIONS

Prepared by

J. B. Blake  
Space Sciences Laboratory  
The Aerospace Corporation

K. L. Hainebach  
Lawrence Livermore Laboratory

D. N. Schramm and J. D. Anglin  
Enrico Fermi Institute  
University of Chicago

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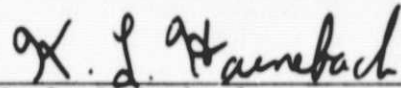
The Ivan A. Getting Laboratories  
THE AEROSPACE CORPORATION  
El Segundo, Calif. 90245

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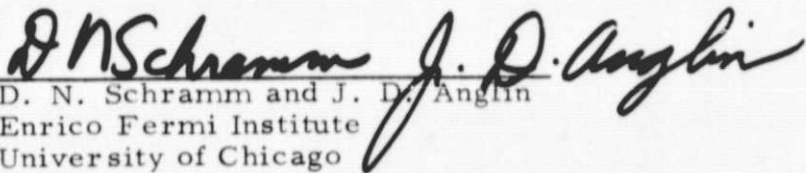
Prepared



B. Blake  
Space Sciences Laboratory  
The Ivan A. Getting Laboratories

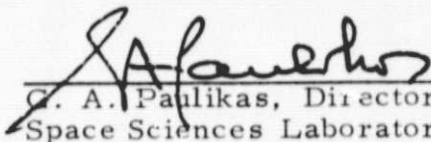


K. L. Haenebach  
Lawrence Livermore Laboratory



D. N. Schramm and J. D. Anglin  
Enrico Fermi Institute  
University of Chicago

Approved



G. A. Paulikas, Director  
Space Sciences Laboratory

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## ABSTRACT

The recent extreme ultraheavy cosmic-ray observations ( $Z \geq 70$ ) are compared with r-process models. A detailed cosmic ray propagation calculation is used to transform the calculated source distributions to those observed at the earth. The r-process production abundances are calculated using different mass formulae and  $\beta$ -rate formulae; an empirical estimate based on the observed solar system abundances is used also. There is the continued strong indication of an r-process dominance in the extreme ultra-heavy cosmic rays. However it is shown that the observed high actinide/Pt ratio in the cosmic rays cannot be fit with the same r-process calculation which also fits the solar system material. This result suggests that the cosmic rays probably undergo some preferential acceleration in addition to the apparent general enrichment in heavy (r-process) material. An estimate also is made of the expected relative abundance of superheavy elements in the cosmic rays if the anomalous heavy xenon in carbonaceous chondrites is due to a fissioning superheavy element.

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## I. INTRODUCTION

Experimental measurements of the extreme ultra-heavy cosmic rays ( $Z \geq 70$ ) are of great interest in the study of nucleosynthesis. Ultra-heavy nuclei are synthesized primarily by neutron capture on either a rapid (r) or slow (s) timescale relative to  $\beta$ -decay rates (Burbidge et al. 1957, Seeger et al. 1965). [See also the discussion of the generalized n-process (Blake and Schramm, 1976)]. By examining the relative abundance of the various ultra-heavy nuclei in cosmic rays, one can judge the importance of each process in the cosmic-ray source. Conventional wisdom attributes the origin of the bulk of the cosmic rays to supernovae and, thus, since r-process nucleosynthesis is believed to occur in supernovae whereas s-process nucleosynthesis is not, the ultra-heavy cosmic rays are expected to be largely r-process in origin (Schramm 1972, Blake and Schramm 1974, Hainebach et al. 1976). The ultra-heavy data currently available (Fowler 1973, Price and Shirk 1975, Price 1976, Fowler et al., 1977) definitely show an r-process character in that the Pt charge group ( $74 \leq Z \leq 79$ ) is substantially more abundant than the Pb charge group ( $80 \leq Z \leq 84$ ) and the actinide nuclei are prominent. The publication of the Skylab results by Price and Shirk (1975) increased by fifty percent the number of ultra-heavy nuclei observed and has the advantage, relative to the other work, of being acquired above the atmosphere, albeit inside the Skylab hull ( $\sim 1 \text{ gm/cm}^2$ ).

The nuclear parameters (mass law,  $\beta$ -rates, fission rates, fission fragment mass distribution) necessary for r-process calculations are not known but must be estimated by theoretical analysis and extrapolation from known nuclei. Examination of nearly pure, and young r-process material in the ultra-heavy cosmic rays (if this is established unquestionably to be the case) can reveal much about the details of the r-process, in particular, and supernovae in general.

In this paper the implications for nucleosynthesis theory are considered of the ultra-heavy cosmic-ray experimental results. First the basic uncertainties in the r-process calculations are discussed. Next various r-process source-abundance distributions are used in a galactic propagation calculation and compared with the Skylab results of Price and Shirk (1975). This is the most complete cosmic-ray propagation calculation in this mass region to date and includes a complete reaction network. It is found that the disparity between the high actinide/platinum ratio in the cosmic rays and the lower ratio in solar system material cannot be explained solely by propagation effects operating on the r-process abundances resulting from any single mass law and  $\beta$ -rate formalism. Preferential acceleration effects are required. These are discussed and comment is made on uncertainties in the solar system abundances of Pb, Bi and the actinides. The importance of improved charge resolution in ultra-heavy cosmic-ray measurements and the question of superheavy element abundances are addressed.

## II. r-PROCESS CALCULATIONS

Because the nuclei involved in an r-process event are far removed from the valley of  $\beta$ -stability (Burbidge et al. 1957, Seeger et al. 1965), the relevant nuclear parameters are not obtained from experimental measurements. Rather, the mass law and  $\beta$ -rates used in r-process calculations are estimated by theoretical analysis and extrapolation from known nuclei.

The Myers and Swiatecki (1966) liquid-drop mass law with the modification to the shell correction given by Myers and Swiatecki (1967) is used in the r-process calculations discussed in this paper. The newer droplet model is available (Myers and Swiatecki 1969, Myers 1976) and has been used extensively in calculations also. However the differences in the calculated abundances as a result of using the droplet model instead of the liquid drop model are small and immaterial for present purposes. The liquid-drop model is used in order that a direct comparison can be made with the previous calculations and discussions (viz. Schramm and Fiset 1973, Blake and Schramm 1974). The next neutron magic number after  $N = 126$  is taken to be  $N = 184$  and the next proton magic number after  $Z = 82$  is taken to be  $Z = 114$  in the liquid-drop mass law. The values of these next magic numbers are not known from experiment and are subject to theoretical debate (cf. Nilsson 1972); this is discussed further below. For nuclei in the actinide region and below, the selection of the next proton magic number to be  $Z = 114$  or  $Z = 126$  is not significant; it is crucial only to a discussion of the r-process synthesis of superheavy elements.

Two modern descriptions of the  $\beta$ -rates are available. One is basically the Fermi theory and has been discussed from the point of view of its r-process applications by Senbetu (1973). The second  $\beta$ -rate formalism is the Gross theory of  $\beta$ -decay (Takahashi and Yamada 1969, Kodama et al. 1970, Takahashi, 1971) and has been described and used

in r-process calculations by Kodama and Takahashi (1975). The Senbetu formalism is a "simple" description which requires an arbitrary  $\log ft$  value and averages overall states without special weighting. The Gross theory includes more of the relevant physics. Because nuclei far from  $\beta$  stability have large  $Q$  values, their decays proceed to many final states. The Gross theory was devised to treat this situation and, instead of dealing with individual transitions, it directly treats the  $\beta$  strength functions. However, it must be emphasized that the nuclei of interest in an r-process are far removed from the region where experimental data presently exists. Any  $\beta$ -rate formalism thus is a large extrapolation into the unknown. It turns out that the major difference in the results of the present calculations between the two different  $\beta$ -rates (for r-process abundances of interest in the present paper) is that the abundance of the actinides relative to the platinum peak is a factor of  $\sim 3$  larger using the Gross theory.

In the Senbetu  $\beta$ -rate formalism, the transition probabilities,  $\log ft$ , are chosen to be some average value for all transitions. The value chosen (usually between 5 and 6.5) does not in any way affect equilibrium r-process calculations since it treats all transitions equally. It is of significance in dynamic calculations (Schramm 1973, Hillebrandt and Takahashi 1976) but even then has little effect on relative abundances in a given mass region. It is possible to have the  $\log ft$  value vary with  $A$  when using the Senbetu formalism (Blake and Schramm 1974). An examination of the  $\beta$ -rates of nuclei near the valley of  $\beta$  stability indicates that  $A \approx 80$  may be a transition point from  $\log ft \approx 5$  to  $\log ft \approx 6.5$ . However, far from the valley of stability, there is no experimental data available to guide the selection. In the case of the Gross theory of beta decay, the  $\log ft$  values are not free parameters.

As indicated above, variations in the mass law for a given description of the  $\beta$ -rates also strongly affects the r-process abundance predictions (Seeger 1967, Mathews and

Viola 1976). A marked effect (for present purposes) arises if the next neutron magic number after  $N = 126$  is  $N = 164$  rather than  $N = 184$ ; such is the case in the mass law of Seeger and Howard (1975). A magic number at  $N = 164$  yields an abundance peak on the valley of  $\beta$ -stability at  $A \approx 250$ , in the region of the heavy actinides. The result is that the heavy actinides would be produced in much greater abundance than the low-mass actinides, the short-lived trans-bismuth nuclei, lead, and bismuth. Mathews and Viola (1976) show the shape of an r-process production curve using the Seeger and Howard (1975, 1976) mass law. Such a production curve is not a good fit to the empirical data. Furthermore, theoretical opinion appears to strongly favor  $N = 184$  as the next magic number after  $N = 126$ . Mathews and Viola (1976) also give an r-process production curve using the empirical mass law of Viola et al. (1974); this mass law gives a production in the actinide region within a factor of two of the platinum peak. The empirical evidence argues strongly against this mass law; actinides would have a much greater abundance relative to the platinum peak than is observed.

The results presented in this paper are from static (constant temperature, constant neutron density) r-process calculations. Because the astrophysical setting of the synthesis of the neutron-rich nuclei attributed to r-process is unknown (cf. Schramm and Norman 1976), a more complex (dynamic) calculation simply adds additional free parameters. Although the dynamic calculations are more "realistic" and might eventually permit identification of the r-process site, such calculations do not give a convenient focus on the underlying nuclear physics because of the substantial additional complication caused by the hydrodynamic parameterization. It has been shown (Schramm 1973) that the general character of the smoothed dynamic solution is similar to that obtained with a straightforward static calculation in which the temperature and neutron density are chosen in such a way as to produce the observed abundance peak locations. A good fit

occurred in the present calculations for  $T_9 = 1.8$  and  $\log_{10}$  neutron density = 28. In a dynamic calculation the neutron flux decreases with time (Schramm 1973, Hillebrandt and Takahashi 1976, Schramm and Norman 1976). However, if all the actinides are synthesized in a given event, whether the r-process path terminates due to neutron induced fission or neutron exhaustion is irrelevant to the relative actinide abundances. It is possible for an r-process event to terminate due to neutron exhaustion just as the actinides begin to be produced and thus yield, for example, a Th/Pt ratio of arbitrary value between the standard cyclic solution value and zero. However, the observational data gives such ratios as Pu/Th, U/Th, actinides/Pt, actinides/Bi which suggest that the relative r-process production abundances in the actinide region are quite uniform. This would not be the case if the r-process terminates just as the actinide region is reached.

In addition to the above theoretical calculations, a set of initial r-process abundances implied by the present solar system r-process abundances have been generated. Specifically, these are derived from the present-day abundances of  $^{209}\text{Bi}$  (from the decay of  $^{237}\text{Np}$ ),  $^{232}\text{Th}$ ,  $^{235}\text{U}$ , and  $^{238}\text{U}$  using a constant rate-of-synthesis model for Galactic chemical evolution, and from an assumption of equal initial abundances of all even nuclei (the  $^{232}\text{Th}$  and  $^{238}\text{U}$  chains), and equal initial abundances of all odd nuclei (the  $^{237}\text{Np}$  and  $^{235}\text{U}$  chains).

### III. PROPAGATION CALCULATIONS

It is necessary to account for alteration of the composition of the cosmic rays due to collisions with ambient particles in the interstellar medium and due to radioactive decay. The present calculations depend largely on the semi-empirical formulae for nuclear breakup reaction cross sections given by Silberberg and Tsao (1973a,b). However, for fission and spallation of trans-bismuth nuclei, the fission-corrected semi-empirical formulae of Schramm (1972) have been used. For peripheral reactions on trans-bismuth nuclei other than  $^{238}\text{U}$ , the cross sections used are those of the corresponding reactions for  $^{238}\text{U}$ , but scaled by the relevant total inelastic cross sections and fissilities. Total inelastic loss cross sections were taken from Kirkby and Link (1966).

A steady-state model of cosmic ray transport through exponentially-weighted slab distributions was used for the propagation calculations discussed in this paper. Only energy independent exponential path length distributions were considered and no attempt was made to reproduce the secondary to primary ratios, declining with increasing energy, which have been observed in the light, medium, and heavy cosmic rays (Juliusson 1974). The effects of energy loss are taken into account by assuming stopping powers appropriate to a neutral gas with solar system composition. The resulting model is equivalent to the leaky box model of Gloeckler and Jokipii (1969) with a leakage time for particles from the Galaxy inversely proportional to velocity.

A total of 177 species with  $A \geq 170$  were included in the present calculations. Nuclei with alpha, beta, and fission decay modes were included if  $T_{1/2} \geq 100$  yr. Pure electron-capture nuclei were included if  $T_{1/2}(\text{EC}) \geq 10$  h and  $T_{1/2}(\beta^+) \geq 100$  yr. The cross sections needed to treat electron capture nuclei were calculated using the Brinkman and Kramers (1930) formula for non-radiative capture and the formula given by Bohr

(1948) for electron stripping. One-electron capture cross-sections in the charge region of interest in this paper are large at all energies likely to be of interest in the near future. Thus most of the electron capture species in this mass region should decay during propagation unless they have half-lives comparable to or longer than the age of the cosmic rays. This happenstance greatly reduces the number of electron capture nuclei that need to be included in a detailed study of cosmic-ray propagation in the Pt region and above.

The cross sections of all possible reaction channels were added together in estimating the interaction length for one species going to another species. The mean interstellar density enters the calculation in relating decay half-lives to interaction lengths in the interstellar gas. One of the unique features of the present calculational method lies in the diagonalization of the interaction matrix. This allows an accurate treatment of fast decays and all higher order production modes within the pathlength step chosen for the integration of the partial differential equations describing production and loss of each nucleus along its corresponding energy-loss characteristic. This diagonalization procedure is relatively simple to carry out because the interaction matrix is almost upper triangular when the nuclei are ordered in terms of increasing  $A$  and decreasing  $Z$ . This allows the eigenrow and column vectors to be calculated by straightforward Gaussian elimination.

The source spectra used in this calculation were of the form  $Q_i(E + E_0)^{-2.6}$  where  $Q_i$  is the relative abundance of species  $i$ ;  $E$  is kinetic energy/nucleon; and  $E_0 = 400$  MeV/nucleon. This form of the source spectra, when modulated, produces a good fit to the observed energy spectra of light and medium cosmic ray nuclei (Garcia-Munoz et al. 1975b).

The recent studies of the age of the cosmic rays, using  $^{10}\text{Be}$  (Garcia-Munoz et al. 1975a, 1977a) led to the parameters used in the present propagation calculations: an exponential path-length distribution with a  $5 \text{ g/cm}^2$  (of hydrogen) leakage mean-free-path



(6 g/cm<sup>2</sup> including interstellar helium). Cosmic ray propagation calculations frequently assume an interstellar matter density of  $\approx 1$  atom/cm<sup>-3</sup>. However, Garcia-Munoz et al. (1975a, 1977a) found a much lower value,  $\approx 0.2$  atoms/cm<sup>-3</sup>, giving a leakage lifetime of  $2 \times 10^7$  yr. We have done propagation calculations using both 1 atom/cm<sup>-3</sup> and 0.1 atom/cm<sup>3</sup> and present the latter results in this paper as being more consistent with the results of Garcia-Munoz et al. (1975a, 1977a). This lower density leads to a cosmic ray leakage age of  $3.2 \times 10^7$  yr for a velocity of  $\beta = 1$ .

#### IV. RESULTS

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The available experimental data and the results of the propagation calculation done with several initial relative abundances are shown in Table 1. The statement that the experimental data strongly favors an r-process source can be seen explicitly. The high actinide relative abundance and the presence of trans-uranics cannot result from the acceleration of solar-system material. Furthermore, it can be seen that the Gross theory  $\beta$ -rates yield a reasonable fit to the measured U/Pt and Trans-U/Pt ratios, especially to the Skylab data. The Senbetu  $\beta$ -rates yield U/Pt and Trans-U/Pt ratios approximately a factor of three lower than the Skylab results.

In Table 2, the solar system abundances of  $^{232}\text{Th}$ ,  $^{235}\text{U}$ , and  $^{238}\text{U}$  (Cameron 1973) are presented along with the calculated r-process abundances of these isotopes using the two  $\beta$ -rate formalisms. In order to test the consistency of the calculated abundances of  $^{232}\text{Th}$ ,  $^{235}\text{U}$ , and  $^{238}\text{U}$ , an extreme model for galactic evolution was taken in which it was assumed that all r-process elements were created in one burst of nucleosynthesis  $20 \times 10^9$  y ago, i.e. the Hubble age for the Universe. This assumption assures that the maximum amount of actinides will have decayed, and gives the lowest possible actinides/Pt ratios at the time of formation of the solar system. One sees that the aged Senbetu abundances are smaller than Cameron's values for solar system abundances, indicating that it is necessary to construct a less extreme galactic evolution model which agrees with the presently observed abundances of these elements (cf. Hainebach and Schramm 1977). However, after aging the Gross-theory abundances, the resulting  $^{232}\text{Th}$  abundance is still a factor of  $\sim 2$  higher than its solar system abundance. In addition, the Gross theory abundance of the  $^{209}\text{Bi}$  decay chain is also a factor of  $\sim 3$  higher than the solar system abundance of  $^{209}\text{Bi}$ .

TABLE 1

	Pb/Pt	U/Pt	Trans-U/Pt
Balloon data (pre-1973) 159 events (Fowler 1973)	$0.4 \pm 0.1$	$0.3 \pm 0.1$	$0.2 \pm 0.2$
Skylab ~ 100 events (Price 1976)	0.6	$0.12^{\S}$	$0.09^{\S}$
r-process with Senbetu $\beta$ -rates: propagated*	500 MeV/n 1000 MeV/n	$0.21 (0.3)^{\dagger}$ $0.23 (0.4)$	0.03 0.04
r-process with gross theory $\beta$ -rates: propagated*	500 MeV/n 1000 MeV/n	0.5 (0.7) 0.5 (0.7)	0.07 0.09
Solar system (Cameron, 1973) propagated*	500 MeV/n 1000 MeV/n	0.8 (1.2) 1.0 (1.5)	0.000025 0.000004
Implied Solar System r-process	500 MeV/n 1000 MeV/n	0.21 (0.3) 0.22 (0.3)	0.03 0.04

Pt  $\equiv 74 \leq Z \leq 79$ ; Pb  $\equiv 80 \leq Z \leq 84$ ; U  $\equiv 90 \leq Z \leq 92$ ;

Trans-U  $\equiv Z \geq 93$ ; except for Fowler (1973) results

where Pt  $\equiv 75 \leq Z \leq 79$ ; Pb  $\equiv 80 \leq Z \leq 84$ ; U  $\equiv 90 \leq Z \leq 94$   
and Trans-U  $\equiv Z \geq 95$ .

\* Exponential path-length distribution with leakage mean free path 5.0  
g/cm<sup>2</sup> of hydrogen and interstellar density of 0.1 atoms/cm<sup>3</sup>. Mean  
age thus is  $3.2 \times 10^7$  yrs for velocity  $\beta = 1$ .

<sup>†</sup> MeV/nucleon

<sup>‡</sup> Values in parenthesis reflect preferential acceleration (discussed in the text).

<sup>§</sup> 4 U events, 3 Trans-U events.

TABLE 2

## r-Process Actinide Abundances

Progenitor chain*	Summed initial solar abundances †	Initial r-process abundances Senbetu $\beta$ -rates ‡	Senbetu $\beta$ -rates, aged $15 \times 10^9$ yr	Initial r-process abundances $\pm$ gross theory †	Gross theory $\beta$ -rates aged $15 \times 10^9$ yr ‡
$^{209}\text{B}$	0.143	0.140	0.140	0.605	0.605
$^{232}\text{Th}$	0.058	0.053	0.025	0.23	0.11
$^{235}\text{U}$	0.0063	0.048	$1.8 \times 10^{-8}$	0.19	$7.2 \times 10^{-8}$
$^{238}\text{U}$	0.020	0.033	0.0032	0.14	0.013

Progenitor chain*	Initial r-process abundances Senbetu $\beta$ -rates ‡	Senbetu $\beta$ -rates, aged $15 \times 10^9$ yr	Initial r-process abundances $\pm$ gross theory †	Gross theory $\beta$ -rates aged $15 \times 10^9$ yr ‡
$^{209}\text{B}$ , $^{209}\text{B}$ , $^{213}\text{Po}$ , $^{217}\text{At}$ , $^{221}\text{Fr}$ , $^{225}\text{Th}$ , $^{229}\text{Th}$ , $^{233}\text{U}$ , $^{237}\text{Np}$ , $^{241}\text{Am}$ , $^{245}\text{Cm}$ , $^{249}\text{Cf}$ , $^{253}\text{Es}$ , $^{257}\text{Fm}$				
$^{232}\text{Th}$ : $^{232}\text{Th}$ , $^{236}\text{U}$ , $^{240}\text{Pu}$ , $^{244}\text{Pu}$ , $^{248}\text{Cm}$ , $^{252}\text{Cf}$ , $^{256}\text{Fm}$				
$^{235}\text{U}$ : $^{235}\text{U}$ , $^{239}\text{Pu}$ , $^{243}\text{Am}$ , $^{247}\text{Cm}$ , $^{251}\text{Cf}$ , $^{255}\text{Fm}$				
$^{238}\text{U}$ : $^{238}\text{U}$ , $^{242}\text{Pu}$ , $^{246}\text{Cm}$				

\* The chains are:

Some chains may go higher, cf. Schramm and Fowler (1971).

† Cameron (1973):  $\text{Si} = 10^6$ ; values at the time the solar system condensed  $4.6 \times 10^9$  yr ago.‡ Normalized to  $^{195}\text{Pt} = 0.473$ , the solar-system abundance.\*  $15 \times 10^9$  yrs before the solar system condensed, i.e.,  $\sim 20 \times 10^9$  yr before the present, which is the Hubble age.

Thus, it is not possible to construct a model of galactic nucleosynthesis using the Gross theory of  $\beta$ -decay which is consistent with the Cameron (1973) solar system abundances without assuming that both  $^{209}\text{Bi}$  and  $^{232}\text{Th}$  are depleted relative to  $^{238}\text{U}$  by chemical fractionation during the formation of the solar system.

An examination of the meteoritic data collected in Mason (1971) reveals that it is unlikely although not inconceivable that the  $^{232}\text{Th}$  abundance adopted by Cameron (1973) is in error by as much as a factor of 2. The solar  $^{232}\text{Th}$  abundance was recently reduced by a factor of four and now agrees with the present meteoritic abundance as a result of a new value for the Th oscillator strength derived from laboratory measurements (Anderson and Petkov 1975). The meteoritic abundances of Bi and U are better determined. Both elements have abundance variations of less than 10% in C1 chondrites (cf., Krähenbuhl et al. 1973). Bi is more volatile than Th or U as evidenced by the fact that it is depleted in C2 chondrites by a factor of about 2. However, the degree of chemical fractionation of Bi and Th required to make the level of actinides produced in the Gross-theory calculation consistent with the solar system abundances of these elements is unlikely.

In order to explain the U/Pt and Trans-U/Pt ratios observed in the ultra-heavy cosmic rays, one needs a total actinide abundance similar to that predicted by the present static r-process calculation using the Gross-theory  $\beta$ -rates (see Figure 1). Increasing the odd to even A abundances in the actinide region while maintaining the same total actinide abundance eases the  $^{232}\text{Th}$  problem but makes the required fractionation of  $^{209}\text{Bi}$  even greater. It is possible to reduce the  $^{232}\text{Th}/^{238}\text{U}$  production ratio about a factor 2 to obtain agreement with the solar system ratio but one is still left with too much  $^{209}\text{Bi}$ . The basic problem is that there are more long-lived actinides with odd A than even A

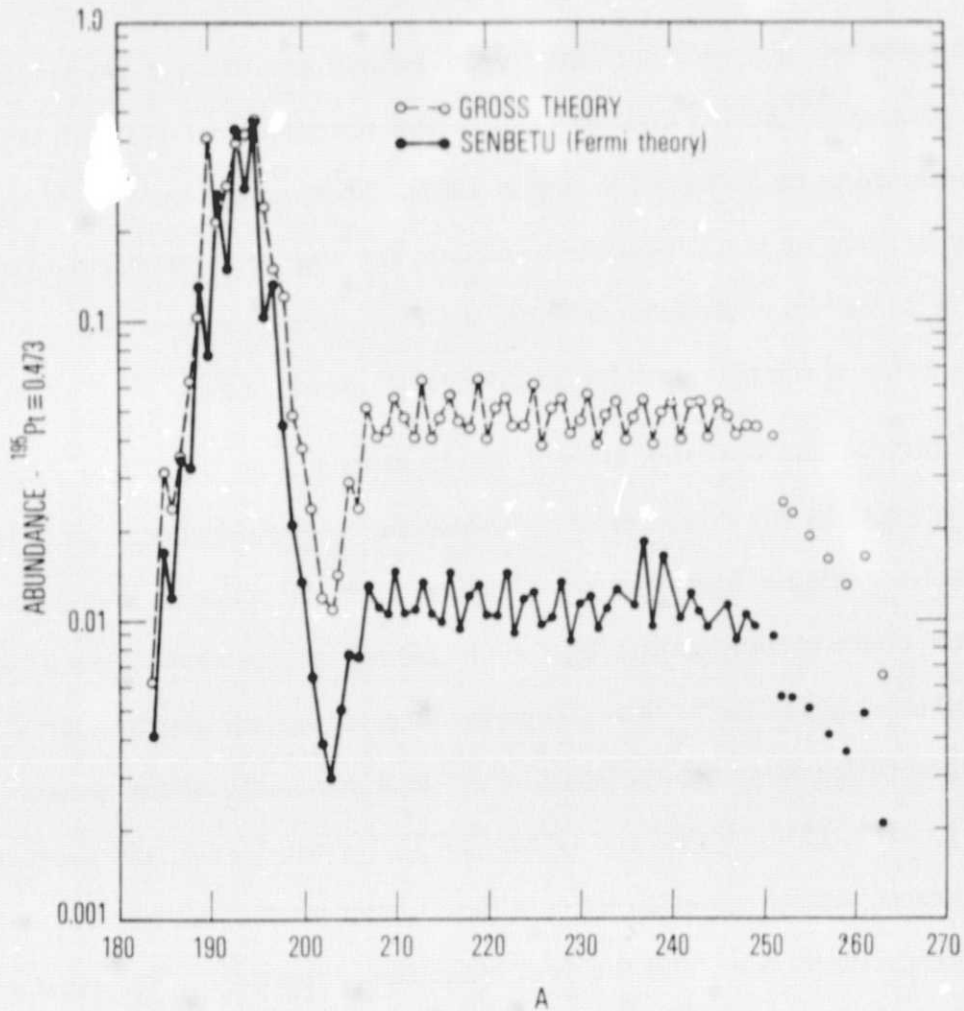


Fig. 1. The calculated r-process abundances for  $A > 180$  are shown using both the Senbetu  $\beta$ -rates and the Gross theory  $\beta$ -rates. The liquid drop mass model was used and a temperature of  $T_9 = 1.8$  and  $\log$  neutron density = 28 (which fits the observed solar-system peak platinum at  $A = 195$ ). Adjacent values of  $A$  are connected by lines to guide the eye except at high atomic weight where spontaneous fission (on the valley of stability) eliminates some of the adjacent mass numbers.

contributing to the abundances of the ultra-heavy cosmic rays, and for published estimates of odd to even production ratios it is not chronologically possible to increase the production abundance of actinides sufficiently to explain the cosmic ray abundances without over-producing  $^{209}\text{Bi}$  (further details on the chronological restrictions on r-process abundances are to be discussed in Anglin 1977). Thus, unless the cosmic ray data is systematically in error, it is not possible to explain the high actinide abundance in the cosmic rays and solar system actinide abundances with the same r-process. One must look for another explanation of the high actinide abundance of cosmic rays.

One possibility is that selection effects, which depend upon the atomic properties of the elements, operate at the source prior to and/or during acceleration. An argument against such selection effects operating on ultra-heavy cosmic rays is that while they appear to occur for elements lighter than Si, the cosmic ray source abundances of Si, Ca, Fe, and Ni are remarkably similar to those found in the solar system (Garcia-Munoz et al. 1977b). However, selection effects can't be ruled out as a possibility at the present time. Kristiansson (1971, 1972, 1974), Cassé and Goret (1973), Havnes (1973), and Cassé et al. (1975) have considered various selection effects based upon nuclear charge  $Z$ , electron-impact ionization cross-section  $\sigma$ , and the first ionization potential  $I$ . The most recent paper, Cassé et al. (1975), examines all three possibilities. Their analysis suggests that element selection dependent on ionization potential is the most reasonable, with overabundance increasing with decreasing ionization potential. Values for  $I$  for the elements of major interest in the ultra-heavy cosmic rays data discussed here are given in Table 3.

It is clear from the run of values in Table 3 that the actinides would be favored relative to the elements in the platinum peak if the abundances have the proposed dependence upon ionization potential. In order to quantify the result somewhat, the data

TABLE 3

## Ionization Potential of Selected Elements

Element	Ionization Potential, eV
Os	8.70
Pt	9.00
Au	9.22
Pb	7.42
Th	7.50
U	6.19
Pu	5.71

From the compilation of Kaye and Laby (1973) .



presented by Cassé et al. (1975), (their Figure 1-c relating relative abundance to ionization potential) have been fit with a straight line (on a semi-log plot) for  $5 \text{ eV} \leq I \leq 15 \text{ eV}$ . (The data for H, Ne, and He were not used.) The straight-line fit predicts the following enhancements for U and Pb relative to Pt:

U/Pt enhanced by a factor of 2.5,

Pb/Pt enhanced by a factor 1.5.

It is interesting to note that if such ionization-potential-dependent enhancements actually occur, using the results of either the r-process calculations with the Senbetu  $\beta$ -rates or the implied solar system r-process abundances after propagation yield a good fit to the cosmic ray (except perhaps Pb/Pt, but see below). However, the Gross theory  $\beta$ -rates now yield too high an actinide/Pt ratio. In this regard, note that the solar system abundances of stable r-process elements are fit better using the Senbetu  $\beta$ -rates than the Gross theory  $\beta$ -rates (Blake and Schramm, 1974). It is clear that an understanding of preferential acceleration or other selection effects is crucial to further progress in relating observations to theoretical r-process calculations.

Note that the Pb/Pt ratio would be increased by preferential acceleration as discussed above. Preferential acceleration thus would not be able to alter a solar system composition source into an enriched Pt peak relative to Pb, but in fact would deplete Pt relative to Pb. One further point to remember is that preferential acceleration of the radioactive nuclei between Bi and Th would further increase the Pb/Pt ratio since these radioactive nuclei will decay to the Pb peak, thus bringing the Senbetu  $\beta$ -rates r-process abundances, with preferential acceleration, into better agreement with the Skylab data.

The possibility that r-process events differ in their production ratios and that the cosmic ray source and solar system material each comprise different distributions of r-process events should be considered. However, the sharpness of the solar system r-process peaks, e.g. the  $^{195}\text{Pt}$  peak, argues against this: if the location in mass number of the peaks from various events differed much, the composite peak would be much broader than observed. One might instead propose that the cosmic ray source consists of a few peculiar supernovae (e.g. Chevalier's suggestion, discussed below) which contribute little to Galactic chemical evolution, but in which the r-process environment persists longer and puts more material into heavier nuclides, like the actinides, than does the more common (in this model) r-process. This rare r-process would be the ordinary cyclic solution r-process (Seeger et al. 1965) in which material eventually reaches some value of A where neutron-induced fission occurs, the fission fragments thereafter serving as r-process seed nuclei. In order to account for the high cosmic ray actinide/Pt ratio, it would have to be assumed that the usual theoretical cyclic r-process is actually rare in nature and that solar system material came from a non-cyclic r-process, which gave a lower actinide/Pt ratio. But there is evidence that solar system material is the result of a cyclic r-process, viz. the abundance "hump" in the rare earth region which could be the result of fission of material at the N=184 magic number (Schramm and Fowler 1971). Thus while a non-standard r-process might explain (in an ad hoc fashion) the high cosmic ray actinide/Pt ratio, such an explanation, upon examination, is not very promising.

It is possible also that uncertainties in the measurements might have significantly affected the published results. Price and Shirk (1975) give a median charge resolution of  $\Delta Z = 2.5$ . They take Au ( $Z = 79$ ) as the upper bound of the platinum peak and Hg ( $A = 80$ )

as the lower bound of the lead peak. The r-process is the major source of Au and even of Hg. Thus substantial amounts of r-process material may well be assigned to the lead peak; the also Pt itself has  $Z = 78$ . The lead peak is overwhelmingly Pb ( $Z = 82$ ) and Bi ( $Z = 83$ ); thus little actual lead-peak (i.e., s-process) material would be expected to be misassigned to the platinum peak.

The r-process indirectly produces substantial amounts of lead-peak material. As the actinide production increases, the lead peak does also, due to the fact that the lead peak (as produced in the r-process) largely arises from the rapid decay of nuclei with  $84 \leq Z \leq 89$ , and not from the direct production of Pb and Bi. The dilemma, that a substantial lead peak can be due either to enhanced actinide abundances or to the presence of s-process nuclei, can be experimentally addressed with a good charge-resolution measurement (UK-6 and HEAO-C) able to separate Pb from Bi. For nuclei created in the r-process  $\sim 10^7$  yr prior to observation,  $\text{Bi/Pb} \approx 0.5$ , whereas in the solar system (Cameron 1973),  $\text{Bi/Pb} \approx 0.035$ .

Some s-process material, originally present in the envelope of supernovae, would be expected in the ultra-heavy cosmic rays, but the ratio r/s is expected to be of the order of 10 or 20 (Hainebach et al. 1976). Chevalier (1976) has suggested that heavy element acceleration occurs in faint supernovae which result when a star, which would have been a normal type II supernovae, loses its envelope before exploding. If such is the case, the r/s ratio might be even larger.

As mentioned in Section III, the leakage mean free path of  $5\text{g/cm}^2$  of interstellar hydrogen and interstellar density of  $0.1 \text{ atoms/cm}^3$  chosen for our calculations lead to a cosmic ray leakage lifetime of  $3.2 \times 10^7$  yr for light nuclei. The actinides however (as opposed to light nuclei like  $^{10}\text{Be}$ ) have a destruction mean free path of  $\sim 1 \text{ gm/cm}^2$ , and

therefore a smaller mean age, perhaps  $5 \times 10^6$  yr. For this reason, the actinides are not particularly useful cosmic-ray chronometers in that they do not give the leakage lifetime of the bulk of the cosmic rays. If the interstellar density were as high as  $1 \text{ atom/cm}^3$ , the actinide lifetime would be very short indeed.

There is evidence that a truncated exponential path length distribution provides a better fit to the abundances of the secondaries from Li to Mn than does a pure exponential (Garcia-Munoz et al., 1977b). The effect of this change is to increase the mean age of the observed cosmic rays and the relative abundance of secondaries to primaries given a fixed average path-length. In the ultra-heavy cosmic rays, such a path-length distribution would result in a lower actinide/Pt ratio than otherwise and make the observed high ratio even more outstanding in the source.

## V. SUPERHEAVY ELEMENTS

The ultra-heavy cosmic ray experiments have detected no superheavy elements (Fowler 1973; Price and Shirk 1975). However, these limits on the superheavy flux are not stringent.

Recently Anders and colleagues (Lewis et al. 1975; Anders et al. 1975) have carried out a beautiful set of experiments studying the rare gases, in particular the excess heavy xenon isotopes, in separate fractions of the Allende meteorite. They argue that the excess xenon may be the product of a fissioning heavy nucleus and, since this putative progenitor is shown to be a volatile, actinides are excluded. The results of the detailed analysis of Anders et al. (1975) suggest that the superheavy progenitor had a nuclear charge of 113, 114 or 115 and that the xenon cannot be due to the presence of primordial grains. If the xenon is indeed due to a fissioning nucleus, the half-life must be such that a significant amount could survive the time interval between the last addition of newly synthesized nuclei to the solar system material, and the onset of xenon retention. Cosmochronological studies show this interval,  $\Delta$ , to be  $\sim 2 \times 10^8$  yr (Schramm 1974).

The observations of Lewis et al. (1975) and Anders et al. (1975), and the assumption that the xenon is due to a superheavy progenitor, may be used to estimate the expected flux of superheavy cosmic rays relative to the actinide cosmic rays. At the time of xenon retention, the abundance of the superheavy (Sh) relative to  $^{238}\text{U}$  was (Anders et al. 1975)

$$\frac{\text{Sh}}{^{238}\text{U}} \sim 6 \times 10^{-4}. \quad (1)$$

It is also known from other meteoritic studies (cf. Lewis et al. 1975 and earlier refs. therein) that

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$$\frac{{}^{244}\text{Pu}}{{}^{238}\text{U}} \approx 3 \times 10^{-2} \quad (2)$$

at the time of xenon retention. Since the value of  $\Delta$  is  $\sim 2 \times 10^8$  yr., one has, from equations (1) and (2), and the usual prescription for radioactive decay:

$$1.8 \times 10^{-2} = \frac{\text{Sh}}{{}^{244}\text{Pu}} \Big|_{\text{Xe retention}} \quad (3)$$

$$= \frac{\text{Sh}}{{}^{244}\text{Pu}} \Big|_{\text{production}} \times \left[ \exp -2 \times 10^8 \left( \frac{1}{\tau_{\text{Sh}}} - \frac{1}{1.2 \times 10^8} \right) \right]$$

where  $1.2 \times 10^8$  is the mean life of  ${}^{244}\text{Pu}$ , and  $\tau_{\text{Sh}}$  is the mean life of the fissioning superheavy progenitor of the xenon.

Studies of the nucleosynthesis of superheavies in the r-process indicates that the ratio  $\frac{\text{Sh}}{\text{Pu}} \Big|_{\text{production}}$  is certainly not greater than unity (Schramm and Fiset 1973). Using this upper limit for the production ratio, equation (3) yields (cf. Schramm 1971)

$$\tau_{\text{Sh}} \geq 3.5 \times 10^7 \text{ yr.} \quad (4)$$

If the ultra-heavy cosmic ray age is as large as the  ${}^{10}\text{Be}$  age (Garcia-Munoz et al. 1975a), viz.  $2 \times 10^7$  yr, the calculated r-process abundances of Blake and Schramm (1974) plus expression (4) give

$$\text{Sh} : \text{Trans-U} : \text{U} = (\leq 0.06) : 0.45 : 1. \quad (5)$$

Many unsuccessful attempts have been made to find superheavy elements in solar system material (cf. Hermann 1974). The negative result is taken to indicate that the mean life of the superheavy progenitor of the fission gas is not greater than that of  $^{235}\text{U}$ , i.e.,  $1.02 \times 10^9$  yr. Using this upper limit yield, from equation (3):

$$\frac{\text{Sh}}{\text{Pu}} \geq 4.1 \times 10^{-3} \text{ production} \quad (6)$$

Again, taking the ultra-heavy cosmic-ray age as  $2 \times 10^7$  yr gives

$$\text{Sh} : \text{Trans-U} : \text{U} = (\geq 0.0004) : 0.45 : 1 \quad (7)$$

However, the age of the ultraheavy cosmic rays may be much less than the  $^{10}\text{Be}$  cosmic ray age because of the large cross section for fission during propagation. If the ultraheavy cosmic ray age were only  $5 \times 10^6$  yr, as suggested in Section IV, the abundance ratios, and upper and lower limits on superheavies become:

$$\text{Sh} : \text{Trans-U} : \text{U} = (0.0005 - 0.10) : 0.75 : 1. \quad (8)$$

If the ultraheavy cosmic ray age were as little as  $10^6$  yr, this becomes:

$$\text{Sh} : \text{Trans - U} : \text{U} = (0.0005 - 0.12) : 1.2 : 1. \quad (9)$$

These results may be summarized as follows: Using the Allende analysis of Anders et al. (1975) plus reasonable but broad limits on the possible production abundance and

lifetime of superheavy nuclei (Schramm and Fowler 1971, Schramm and Fiset 1973) gives a predicted superheavy element flux in the cosmic rays of

$$0.0004 \leq \frac{\text{Sh}}{\text{U}} \leq 0.12. \quad (10)$$

Recently Flerov (1977) has summarized the results of searches for superheavy elements in carbonaceous chondrites and water samples from hot springs. Multiple neutron emission has been detected which cannot be explained by the spontaneous fission of the  $^{238}\text{U}$  known to be in the sample ( $^{238}\text{U}$  is the only naturally occurring nucleus with a significant probability of spontaneous fission decay). These results may be evidence for the presence of a long-lived  $[\tau_{1/2}(\text{Sh}) \sim \tau_{1/2}(^{235}\text{U})]$  superheavy nucleus with a concentration in the meteorite of the order of  $10^{-14}$  g/g. Of course these results need to be verified. However one can say already that if superheavy elements with lifetimes near that of  $^{235}\text{U}$  are found to exist, and these nuclei are nucleosynthetically produced in conjunction with the actinides, then superheavy nuclei eventually should be found in the cosmic rays.



## VI. CONCLUSIONS

An analysis of the ultraheavy cosmic-ray data, especially those collected during the Skylab mission, yields the following results:

1. For the nuclei in the platinum peak and above, a basically r-process source is required as noted earlier (Fowler 1973, Price and Shirk 1975).

2. The disparity between the high actinide/platinum ratio in the (propagated) cosmic rays and the lower ratio observed in solar system material cannot be explained by the use of a single  $\beta$ -rate formalism and mass law in an r-process calculation unless another process, such as preferential acceleration, enhances this ratio in the cosmic ray source.

3. Using reasonable but broad limits on the possible production abundance and lifetime of superheavy nuclei and results of the analysis of the Allende meteorite by Anders et al. (1975) gives a predicted superheavy element flux in the cosmic rays of  $0.0004 \leq \frac{\text{Sh}}{\text{U}} \leq 0.12$ .

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