THE MISSING HEAVY ELEMENTS*

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ABSTRACT. With a view to analysing the possible causes of (1) absence of natural elements having atomic number $Z > 92$ and (2) non-occurence of isotopes of the $(4n+1)$ type among the natural elements, the known isotopes of 93 N and of the transuranium elements from 93 Np to 96 Cm are compiled from the recent works of Seaborg and others. A plot of the Heisenberg energy surfaces for the observed α-emitting nuclei on a neutron-proton diagram reveals the probable existence of an energy-high in the region of lower isotopes of N, Np and Pu. Natural extinction of these isotopes by short-life radioactive α-decay is, therefore, considered possible. The mean life of spontaneous fission of heavy isotopes is also calculated from Turner's extension of Bohr and Wheeler's theory of nuclear fission. The results show that the elements 95 Am and higher would have too short mean fission life to survive in nature. The higher isotopes of 93 Np and 94 Pu present difficulties as neither their radioative decay period nor spontaneous fission life seems to be short enough to account for their non-existence. The processes of production of these isotopes by the observed nuclear reactions in the laboratory are compared with the natural processes of formation of heavy elements in the interior of dense stars as postulated by Weissäcker, Mayer and Teller and others. It is pointed out that, unlike in the laboratory reactions, the heavy elements are probably formed in the stellar processes in very high states of excitation favourable for spontaneous fission and subsequent neutron evaporations from the fission fragments which can lead only to lower isotopes of medium heavy elements. This seems to offer at least a qualitative explanation of the absence of heavy isotopes of Np and Pu in nature, the lower isotopes of which are, as already shown, highly unstable against radioactive decay. The same reason makes it possible that the nuclear reactions which lead to the formation of $(4n+1)$ nuclei in the laboratory cannot take place in the stellar processes on account of serious competition from spontaneous fission processes of the highly excited heavy nuclei formed in these processes.

Natural radioactive elements fall under the three well-known families, U-Ra, Th and Ac-series, which all begin with a long lived heavy parent element, pass through comparatively short-lived intermediate products and ultimately terminate in a stable end product, which is an isotope of Pb. The heaviest element occurring in the series is $_{92}U^{238}$, the parent of the U-Ra-series, having an α-decay mean half-life $T \sim 4.5 \times 10^9$ years. $_{91}Pa^{231}$, the parent of the Ac-serles, is geneologically connected to a rarer isotope of uranium, U^{235}($T \sim 7.1 \times 10^8$ years) through an α- and β^--disintegration. The Th-series originates from $_{90}Th^{232}$ itself possessing a mean half-life $\sim 1.39 \times 10^{10}$ years. The nuclear masses occurring in the three-series are of the $(4n+2)$, $4n$ and $(4n+3)$ type respectively. The three series run nearly parallel courses in many respects in their decay processes and

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altogether 42 radioactive nuclei occur under them. There are, however, two outstanding features of these nuclei, the significance of which is not yet clearly understood, namely, (1) elements with atomic number (Z) higher than 92 do not occur, (2) no member of a radioactive series of the type $(4n+1)$ occurs among the natural isotopes. Attempts have been made by Turner (1945), Feather (1944-46) and others to analyse the possible cause of absence of elements higher than $Z=92$, as being either very high susceptility to spontaneous fission or short-life radioactivity of the absent heavy nuclei. Sufficient data regarding radioactive decay of heavy nuclei in or near this region did not exist at that time to lead to any definite conclusion on the point ; but as pointed out by Feather (*loc.cit.*), the possibility of spontaneous fission being the cause of extinction of the heavy elements beyond uranium seemed to be more or less ruled out. Regarding the non-occurrence of the $(4n+1)$-radioactive series, it has been argued that the elements that could be considered as natural starting points of the series are Th^{233}, U^{237}, U^{233} or Np^{237}, and that none of these nuclei are likely to form in nature, as the α- or β^--disintegrations of the corresponding nuclei which would lead to these parent nuclei do not appear to be possible (Stephens, 1948).

The position has changed somewhat since the discovery of the transuranium elements 93Np, 94 Pu, 95 Am 96 Cm in the laboratory and considerable progress in the studies of their properties. Several isotopes of these elements have now been identified by Seaborg and his associates and their radioactive properties carefully investigated. The relevant results are collected in Table I below (Seaborg, 1942 ; 1946). The purpose of this note is to review the present position of the problem in the light of these new data and to examine the possibility of any further progress in its solution.

Np 239 and Pu 239 are produced in the non-fission neutron-capture in U 238 followed by two successive β^--disintegrations (McMillan and Abelson, 1940). The non-fission capture process in U238 shows a resonance effect at the neutron energy $E_n \sim 25$ eV (Meitner, Hahn and Strassmann, 1937). Pu 239 is α-active having a mean half-life of $\sim$24,000 years. The first isotope of Np and Pu to be discovered was, however, 238, by the (d,2n) process in U 238 followed by a β^--disintegration of Np^{238} with 2 days half-life (Seaborg, McMillan, Wahl and Kennedy, 1941). Then the lighter isotope Np^{237} was obtained by Wahl and Seaborg (1942) by the reaction :

$$U^{238}(n,2n)^*U^{237} \xrightarrow[7\ \text{days}]{\beta^-} Np^{237} \xrightarrow[\sim 2.25.10^6 \text{yrs}]{\alpha} (_{91}Pa^{233}) \qquad \ldots \text{(I)}$$

Np^{237} has a long half life of α-decay and degenerates into Pa^{233}. The transplutonium element Am^{241} was produced by bombarding U^{238} with 40-44 MeV He^{4++} nuclei in the Berkeley cyclotron (Seaborg, 1945). Possible reactions involved are :

$$_{92}U^{238} + {}_2He^{4++} \rightarrow {}_{94}Pu^{241} + {}_0n^1$$

$$\downarrow \beta^-$$

$$95\,Am^{241} \xrightarrow[\sim 500 yrs]{\alpha} {}_{93}Np^{237} \xrightarrow{\alpha} Pa^{233} \quad \ldots \quad (II)$$

The highest element $_{96}Cm^{242}$ has also been synthesised with the fast He^{4++} ions by the reaction (α,n) in Pu^{239} which was already obtained by the simple capture of $_0n^1$ in U^{238}. Cm^{242} has probably a short α-decay life of the order of five months. A branch reaction $(\alpha,3n)$ in Pu^{239} also gives Cm^{240} having a still shorter α-half life of $\sim$ 1 month.

TABLE I

Transuranium and uranium isotopes

93 Np	234 K,γ 4.4 d.	235 K 8 mo.	236 β^- 20 hrs	237 2 25 10^6 yrs (*4.08*)	238 β^- 2 d.	239 β^- 2.3 d	231 *53 min.* (6 2)
94 Pu	*232 22 min.* (6.6)	*234 8 hrs.* (6.2)	*236 2.7 yrs* (5.7)	238 50 yrs. (5.5)	339 24,000 yrs. (5.1)	*240 6,000 yrs.* (5.1)	*241* ~*10 yrs.* (5.0)
95 Am			*239* ~*12 hrs* (5.8)	241 500 yrs. (5.5)	*242* ~*400 yrs.* (5.2)		
96 Cm			*238* ~*2.5 hrs.* (6.5)	240 ~ 1 mo (6.3)	242 5 mo. (6 1)		
92 U	*228 9.3 min.* (6 72)	*229 58 min.* (6.42)	*230 20 8 d.* (5.85)	*232 234* 70-30 2.3.10^5 yrs. yrs. ((5.3) 4.75)	235 8.9.10^8 yrs. (4.52)	* 237 β^- 6 8 d.	238 *239 4.5.10^9 β^- 23 yrs. (4 13) min.

(*Italicised data* are from Seaborg and Perlman (1948). All figures in parenthesis denote radiation energy in MeV. All nuclei are α-emitters, except stated otherwise. When a nucleus decays by β^- or γ-emission or by K-electron capture, the emitted radiation is mentioned below the isotope). *Stephens (1948).

With the new data for many artificially produced heavy radioactive isotopes thus available, we may examine afresh the possible reasons why the heavy elements beyond $Z=92$ would be missing in nature. The limits of stability of nuclei against α-emission have been examined by Heisenberg (Solvay Congress, 1933) by plotting the energy surfaces of the radioactive nuclei for α-emission on a neutron-proton ratio diagram of elements. The general treatment of this type gives a reasonable account of known natural radioactive isotopes, the shortlived C′-products falling into a closed region of energy-high. The new data for α-emission from heavy nuclei as far as curium (96) are, therefore, plotted on an extension of the Heisenberg diagram shown in figure 1. The energy values have been taken, wherever

available, from the experimental data published by Seaborg and others (1942, 1946) from time to time and also from the comprehensive review * of data by Seaborg and Perlman (1948). Doubtful energy values not confirmed by more than one sources and which do not agree with an extrapolation of the Geiger-Nuttall curves have not been plotted.

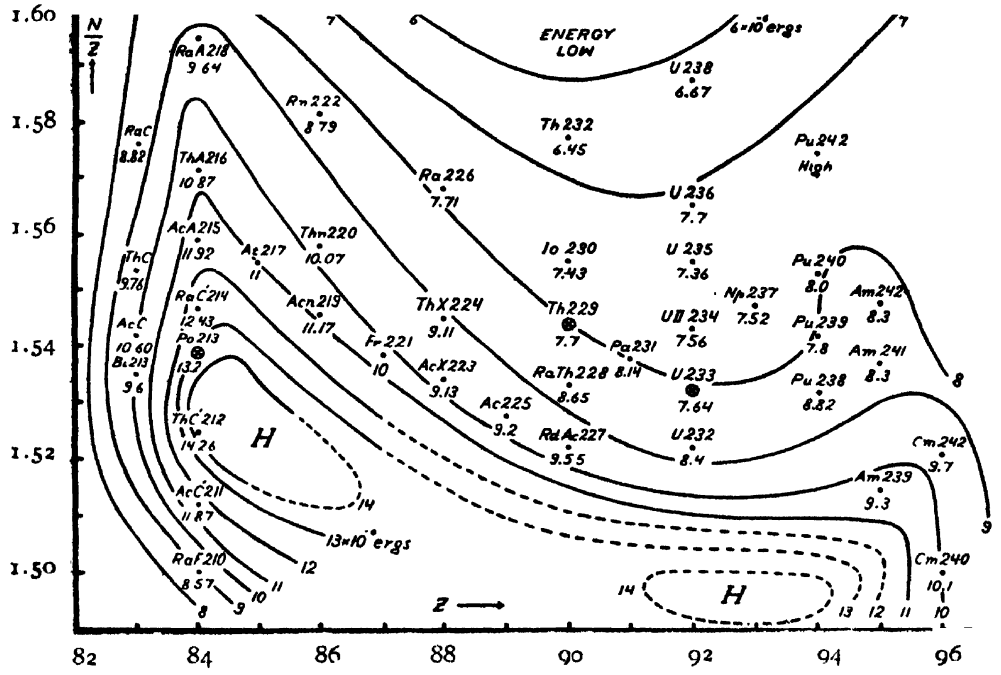

FIG. 1

TABLE II

Isotopes of the $(4n \times 1)$ series. Seaborg (1941).

Nucleus	Radiation	Half-life	Energy of radiation
92 U^{233}	α	1.62×10^5 yrs.	4.80 MeV
90 Th^{229}	α	7000 ± 2000 yrs.	4.45
89 Ac^{225}	α	10.1 ± 0.1 days.	5.80
87 Fr^{221}	α	4.8 ± 0.1 min.	6.30
85 At^{217}	α	0.018 ± 0.002 sec.	7.05
83 Bi^{213}	α(4%)	47 ± 1 min.	6.00
84 Po^{213}	α	very short.	8.30

The rest of the values have been obtained by an approximate extrapolation of the Geiger-Nuttall curves. The values for Th^{229} and Po^{218}, two members of the $(4n+1)$ series, taken from Table II (see discussion at the end)

are seen to fit well in the curves. These two new isotopes, among the well-known radioactive elements, serve well as a check on the new experimental data. The value for U^{233} from the same experimental source does not, however, agree so well. The values extropolated from the G. N.-curve appear to be slightly higher than the experimental energy values in general wherever both the values are available for comparison. But the difference is not so serious as to affect the general run of the Heisenberg curves, which can, therefore, be taken as fairly representative. The position beyond the element 92 is obviously far from satisfactory. But the two isotopes Cm^{242} and Cm^{240} are of great significance as they help to close up the energy curves 10 upwards on the lower side of the N/Z scale. Thus the existence of a second high energy closed region between $Z=91$ and 94 near the bottom of the N/Z scale appears to be fairly certain. We can, therefore, conclude that the lower isotopes of these elements from U to Pu would be highly unstable towards α-decay, and it is no wonder that they do not exist in the earth's crust which formed a few thousand millions years ago.

The higher isotopes of these elements clearly fall on a flat plateau region on the energy diagram. Their α-decay lives are, therefore, not short enough to account for their natural extinction. It is, therefore, necessary to examine the possibility of their decay by alternative mechanisms, like spontaneous fission, neutron-evaporation and β^--emission. The last two alternatives do not probably play any part, as the neutron-evaporation process would lead to lower isotopes which would decay by radioactive emission, while β^--emission would lead to still higher elements which would readily die out by spontaneous fission.

The relative probability of spontaneous fission of heavy isotopes have been considered by Bohr and Wheeler (1939) and by L. A. Turner (1945). While it is clear that the limit of instability of nuclei by spontaneous fission sets in at (Z^2/A) critical ≥ 47.8, Turner has shown that the mean life of spontaneous fission of a heavy nucleus is approximately given by

$$\tau_f \approx \frac{1}{\lambda_f} \approx 10^{-21} \exp.\ [(2\,M\,E_f)^{\frac{1}{2}}.\ a/\hbar]$$

$$\approx k.\ \exp.\ \{A^{7/6}.\ [f(x)]^{\frac{1}{2}}\}$$

where $f(x)=98(1-x)^3/135-11368\ (1-x)^4/34425 + \ldots\ldots\ldots\ldots$,

$x=(Z^2/A)\div(Z^2/A)_{\text{critical}}$, $k=$constant,

$E_f=4\pi r_0^2.\ O.\ A^{2/3}.\ f(x)$,

and the other symbols have their usual meanings. According to this, 94 Pu^{239} should be about 7000 times more spontaneously fissionable than U^{235} which shows spontaneous fission to the extent of $\leq 1\%$ of the radioactive α-decays. Pu^{239} has, therefore, a spontaneous fission life $\tau_f \approx 100.\ 7\times 10^8/7000$ years $\approx 10^7$ years. The ratio of spontaneous fission life of a nucleus to that of

Pu^{239}, denoted by R_f, the relative mean fission life is computed for a number of isotopes by the above formulae and collected in the Table III below.

TABLE III

Spontaneous fission life, R_f, of heavy nuclei relative to Pu 239 [recalculated from $f(x)$].

		R_f			R_f
92 U	235	$7.6.10^3$	94 Pu	244	$5.2.10^4$
	233	$1.1.10^2$		242	$5.9.10^2$
				241	62
93 Np	237	66		229	1.00
				238	0.12
95 Am	243	0.51	96 Cm	240	$7.2.10^{-7}$
	241	0.011		242	$7\ 4.10^{-6}$

R_f = Ratio of spontaneous fission life of a nucleus to that of Pu^{239}.

The above results show that only Am and Cm possess fission mean life low enough to decay completely from the earth's crust in course of geological epochs. Elements higher than Cm will have, of course, still less chance of surviving.

The position of heavy nuclei, therefore, appears to be this : Lower isotopes of elements from 92U to 94 Pu would be extinct through short life α-emission, while the absence of still heavier elements Am and Cm would be accounted for by their high susceptibility to spontaneous fission. The higher isotopes of Np and Pu, which are produced in the laboratory, but do not exist in nature in any appreciable extent are still to be accounted for. Perhaps we can look for an explanation to the probable mechanism of evolution of heavy elements originally in the cosmology as postulated by Weizäcker (1947), Gamow (1948), Mayer and Teller (1948) and others (Haar, 1949).

In the original formation of heavy elements in the interior of very dense and hot stars there must have been heavy nuclei present, because large neutron excess is required for stability of these nuclei in view of strong coulomb interaction of protons. Nuclei with such large neutron excess will not be stable, but would break up by spontaneous fission. The fission products would be unstable and through radioactive changes decay finally into stable nuclei. Mayer and Teller have considered these processes in detail ; they assume that after the fission of the neutron-rich nuclei the residual nuclei will be highly excited. The excess energy of excitation will be first relieved of through " neutron evaporation from the droplet of nuclear

fluid''. If the energy of the nucleus has decreased so far that no more neutrons can evaporate, normal radioactive processes will follow, which will most frequently be β^--processes ($N \xrightarrow{\beta^-} P^+$). The final results will be a stable nucleus—one of the isotopes observed in nature. Starting from the concentrations of neutron-rich nuclei from the stellar interior and making reasonable assumptions as to the excitation energy of the fission products, one can calculate the abundance of the stable isotopes on these lines. Mayer and Teller have found reasonably good agreement between the observed and the calculated values of abundances obtained in this way.

The production of the transuranium elements in the laboratory is probably a process quite analogous to the above natural process, but the degree of energy excitation of the neutron-rich nuclei obtained by the neutron-capture process in U^{238} in the laboratory is much lower than that attainable in the stellar processes. Consequently the subsequent β^--decay process of the compound nucleus formed by the neutron-capture has to suffer little or no competition with the fission process. It is unlikely, therefore, that the neutron-evaporation process can proceed very far, or set in at all. Quite heavy neutron-capture products are, therefore, left behind which, on account of β^--instability settting in quite early, transform gradually to heavier isotopes of Np, Pu, Am and Cm. In the natural evolution process, on the contrary, the fission of very heavy nuclei, followed by neutron-evaporations and β^--decays, probably cannot yield nuclei beyond Th or U.

A few speculative remarks can now be made in the light of the above discussions regarding the non-occurrence of isotopes of the $(4n+1)$-series among the natural radioactive elements. Turner's (1940) suggestion that the starting point of the $(4n+1)$-radioactive series would be ${}_{92}U^{237}$ has not been wholly fulfilled in practice. Extensive experimental researches carried out in connection with the plutonium project have led to the observation of many members of this series and establishment of the sequence of disintegrations almost in complete agreement with Turner's prediction. Only the starting point of the series is found to be the long life α-active Np^{237} which comes out to be genetically related to Pu^{241} and Am^{241} according to the following scheme :

$$ {}_{94}Pu^{241} \xrightarrow{\beta^-} {}_{95}Am^{241} \quad \overset{\alpha}{\searrow} {}_{93}Np^{237} \xrightarrow{\alpha} {}_{91}Pa^{233} \xrightarrow{\beta^-} \text{and so on} $$

The first two members enclosed separately replace U^{237} in Turner's suggestion. The α-emitting nuclei which occur in this series are reproduced (Seaborg, McMillan and others, 1941) in Table II, together with their periods and energy of radiation and have been plotted in the energy diagram, figure 1.

As stated under scheme (II), Pu^{241} is produced from an (α, n)-reaction in U^{238} by a non-fission capture of 40-44 MeV. α-particle. An alternative process of production of Np^{237} is shown in scheme I, which consists of an (n, 2n)—reaction in U^{238} resulting in U^{237}, which is a β^--active body giving rise to Np^{237}. Cross-section for this reaction, as for all (n, 2n)-reactions (Weisskopf, 1950), is extremely small. Therefore, the main reaction giving the isotope Np^{237} in the laboratory must be that under scheme II involving the non-fission capture of an α-particle in U^{238}. But under natural conditions of stellar evolution the non-fission capture will have very little probability, as due to high excitation of the compound nucleus Pu^{242} formed, the fission process would be a serious competitor of the non-fission capture; indeed the spontaneous fission of Pu^{242} may be the main process in nature. This seems to offer at least a qualitative explanation of the non-occurrence of the members of the $(4n + 1)$-series among the natural radioactive elements.

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