NEW ACCELERATORS FOR VERY HEAVY IONS - PRODUCTION OF SUPER-ELEMENTS

Albert Ghiorso

University of California
Lawrence Radiation Laboratory
Berkeley, California

The extension of the periodic system has been a fascinating field of endeavor for many years. The buildup of the elements to atomic number 103, the last of the actinide rare-earth-like series, has considerably enlarged our knowledge of the chemical properties of this series and has produced new insights concerning the nuclear structure of the heavy nuclides. An especially important finding has been the disclosure of the dramatic effects cause by a subshell at 152 neutrons.[1] However, as the atomic number has been increased, the decrease in half-lives and production cross sections has made it increasingly difficult to study these nuclides in any great detail. The purpose of this paper is to give a general picture of the direction of these studies and to indicate some of the requirements imposed on accelerators designed to further this research.

Figure 1 shows the variation in alpha half-life vs neutron number for the elements with even atomic number. Note the prominent peak at 152 neutrons and further note that the alpha half-lives increase again after the dip at 154 neutrons, presumably increasing monotonically until the next neutron shell is reached. The dotted line for element 104 is located where one might logically place it on the basis of the latest data on the preceding elements.[2] The lower limit indicated for the nuclide labeled 104^{260} is already well above this predicted line and thus a question is raised as to the assignment of this 0.3-s spontaneous fission emitter.[3]

The heaviest isotope of element 102 known at the moment, mass 257, has a half-life of 20 s. This value would lead one to predict that 102^{258} should have an alpha half-life in the region of a minute and yet it has not been observed.[2] Fig. 2 indicates the most probably reason for its absence. Plotted is the variation of spontaneous fission half-life with neutron number for elements with even atomic number. The most outstanding characteristic is that a precipitous peak occurs at 152 neutrons as the atomic number is increased. The sharp drop beyond this peak seems to predict a spontaneous fission half-life as short as a millisecond for 102^{258}. Again the 0.3-s activity labeled 104^{260} does not seem to fit in well with a simple empirical extension of the known data for the other heavy nuclides.

If one ignores the data for 104^{260} (and there are other reasons for questioning the assignment of this activity), it is tempting to draw the general conclusion that the rate at which the increase in atomic number decreases the spontaneous fission half-life is really greater than observed and that it is the stabilizing effect of 152 neutrons that partially neutralizes the expected Z^2/A effect. It logically follows from this hypothesis that spontaneous fission will become the predominant mode of decay for the higher Z elements in this region. Since nuclides with an odd number of protons or neutrons are hindered in spontaneous fission decay by factors as great as 10^4 one would expect to find such atoms among the elements with higher Z but it seems probable that the decrease in fission barriers is proceeding so rapidly that the spontaneous fission decay rates for all isotopes may soon become almost instantaneous. Thus production of new elements beyond about atomic number 107 is not very likely if this picture persists.

In 1964, however, Swiatecki and Myers[4] pointed out the possibility that the fission barriers would be raised to rather high levels by the onset of a doubly closed shell at 126 protons and 184 neutrons. These estimates were made by extensions from a semi-empirical mass formula that was found to yield quite reliable data on nuclear masses and deformations of known nuclei. When extrapolated to the region of superheavy nuclei these calculations predicted fission barriers as high as those that assure the stability of the ordinary elements around thorium and uranium. Such exotic nuclei can only be produced by interactions between complex nuclei and it is known that the cross sections for such reactions are proportional to the ratio $\Gamma_n/(\Gamma_n+\Gamma_f)$, where Γ_n and Γ_f are the level widths for neutron emission and fission, respectively. Since this ratio increases as the fission barrier increases, the cross sections to produce nuclides in this hypothetical island of stability should become very large and thus the possibility of producing them is substantial.[5] There is another body of opinion that postulates the next closed proton shell to be at 114 protons. If this turns out to be the case the same general conclusions can be drawn regarding stability but the difficulties in finding suitable reactions for formation of nuclides near 114p and 184n are formidable.

Figure 3 is a Z vs N chart prepared by T. Sikkeland which is intended to represent in a very general way the mountain ranges of stability that might be brought about by shell closures at 126p and 184n. The contours marked with exponents indicate alpha half-lives from 10^{-3} to 10^9 s while those without exponents indicate fission barriers from 2 to 12 MeV. (Note that U^{238} with a spontaneous fission half-life of 10^{16} years has an E_f of 5.8 MeV.) Of course, the alpha half-lives are very dependent on where the line of beta stability is drawn. For an $E_f = 4.0$ MeV the spontaneous fission half-life for an even-even nuclide will be in the neighborhood of seconds so below this contour the nuclides will disappear by spontaneous fission disruption. Above this level on the stability mountain the nuclides will probably disappear by short-lived alpha decay, but in doing so they will change into nuclides of lower atomic number that will decay by spontaneous fission. Thus if these predictions are correct we see that this island of stability is surrounded on all sides by an ocean of spontaneous fission. Such a picture probably rules out production of these superheavy elements by means of the nuclear explosion technique. In this method very neutron-heavy isotopes of a much lighter element are instantaneously formed by the successive amalgamation of a great many neutrons with a light target with subsequent beta decay to a higher Z. It would seem that the beta decay chains would all be interrupted by extremely short-lived spontaneous fission emitters.

What are the best ways of forming these superheavy elements? In the case of those nuclides in the region of 126p and 184n the most promising reactions are those in which the interacting nuclei fuse with subsequent de-excitation by neutron, proton, or γ-emission. The least excitation energy and thus the least fission competition is induced when the projectile and target are of approximately equal mass. As an illustration two systems have been listed with their coulomb barriers V_c, Q values, and corresponding laboratory energy thresholds E_{lab}.

$${}^{96}_{64}\mathrm{Gd}^{160} + {}^{88}_{62}\mathrm{Sm}^{150} \rightarrow {}^{184}126^{310} + \gamma$$

$$\left[\begin{aligned} V_c &= 355 \text{ MeV} \\ Q &= -399 \text{ MeV} \\ E_{lab} &= 773 \text{ MeV } (5.2 \text{ MeV/N}) \end{aligned}\right.$$

$${}^{108}_{72}\mathrm{Hf}^{180} + {}^{78}_{54}\mathrm{Xe}^{132} \rightarrow {}^{184}126^{310} + 2n$$

$$\left[\begin{aligned} V_c &= 348 \text{ MeV} \\ Q &= -419 \text{ MeV} \\ E_{lab} &= 726 \text{ MeV } (5.5 \text{ MeV/N}) \end{aligned}\right.$$

In these cases because of the favorable Q values it is possible to bombard at a C.M. energy that is more than 50 MeV above the barrier and so enhance the cross section.

It is also feasible, though with smaller cross section, to produce the same nuclide by bombarding thorium with krypton ions. Thus:

$${}^{142}_{90}\mathrm{Th}^{232} + {}^{48}_{36}\mathrm{Kr}^{84} \quad {}^{184}126^{310} + 6n$$

$$\begin{aligned} V_c &= 296 \text{ MeV} \\ Q &= -311 \text{ MeV} \\ E_{lab} &= 424 \text{ MeV } (5.0 \text{ MeV/N}) \end{aligned}$$

The cross sections for the above reactions would optimistically seem to fall in the range of 5-50 millibarns and the excitation function half-widths might be in the range of 15-60 MeV. This would imply a maximum usable target thickness of less than 1 mg/cm^2. With a beam of 10^8 ions/s and a cross section of 5 millibarns one could produce 1 dis/s at equilibrium. Of course, if the super heavy nuclide was very long-lived nothing could be observed and more sensitive methods of detection would have to be used to detect it. On the other hand if the stabilization is indeed this high then peripheral nuclides around the island of stability would be observed to decay with measureable half-lives.

The other prominent possibility for doubly-closed shells at 114p and 184n, ${}^{184}114^{298}$, cannot be produced by a fusion reaction followed by neutron or γ-emission only because its high neutron to proton ratio puts it beyond any possible mass combinations. In this case it is hoped that the shell effects will extend over a region wide enough to stabilize nearby nuclides. With stable isotopes the nearest approach would be:

$${}^{82}_{54}\mathrm{Xe}^{136} + {}^{102}_{68}\mathrm{Er}^{170} \rightarrow {}^{184}122^{306} \text{ to produce}$$

a nuclide with 184n

or $${}^{82}_{54}\mathrm{Xe}^{136} + {}^{90}_{60}\mathrm{Nd}^{150} \rightarrow {}^{172}114^{286} \text{ to produce}$$

a nuclide with 114p

Nuclei in the neighborhood of the 114p, 184n double shells may be produced in fusion reactions followed by proton evaporation. No quantitative evaluation of the cross sections for such reactions have been performed in this region of the periodic table. However, for these neutron deficient nuclides the binding energies for protons are less than those for neutrons and hence proton evaporation might compete favorably with neutron evaporation.

There is the remote possibility that ${}^{184}114^{298}$ itself could be formed directly as a primary fission product of the amalgamation of one uranium nucleus with another. It is not clear that such a super nucleation would actually take place but it seems possible that the doubly magic nucleus might be favored in rare cases.

A third possible but rare reaction that might produce nuclides in this region is of the particle transfer type. Thus,

$${}^{102}_{68}\mathrm{Tb}^{170} + {}^{102}_{68}\mathrm{Tb}^{170} \quad {}^{184}114^{298} + (22p\ 20n).$$

This is a grazing reaction in which, possibly, the (22p20n) would be ejected as a cluster. The energetics of this reaction would indicate a bombarding energy of about 6 MeV per nucleon.

There is no possible way that can predict with any reasonable certainty whether these proton and neutron shells will occur and whether the subsequent effects of stabilization will conform to the preceding outline. The only solution must be an experimental one. The requirements for an accelerator that can pioneer this interesting field of research would seem to be the following:

(1) It should accelerate <u>all</u> atoms from Z = 18-92,
(2) It should have a variable energy output from 3-7 MeV/N. with an energy spread of less than 1%,
(3) Though beam currents of 10^8 ions/s are

capable of marginal experiments, it is desirable to have intensities as high as 10^{12} ions/s. Remember however that because of the high mass the very heavy nuclides pose a serious thermal problem. For U^{238} accelerated to 7 MeV/N an average intensity of 10^{12} ions/s would mean about 300 watts dissipated in a total range of about 10 mg/cm^2,

(4) A long duty cycle is imperative for in-beam types of experiments on nuclides with nanosecond half-lives.

During the last two years we have expended a considerable effort to try to decide which type of accelerator was best suited to the above requirements. This study has led us to a completely new accelerator concept which not only will provide suitable heavy ion beams for low energy nuclear chemistry research but will also generate high energy heavy ion beams for an important new area in biomedical research and therapy.

Let us consider the primary problem with conventional acceleration methods when applied to very heavy atoms. This is the difficulty of removing a sufficient number of electrons from the atoms to make efficient acceleration possible. Fig. 4 indicates the relative abundance at each charge state of krypton and xenon for one of the most efficient ion sources available at the present time plotted against ϵ, the charge to mass ratio. It can be seen that at the values of ϵ normally necessary in even large cyclic accelerators, 0.15 or more, the ion output for these moderately heavy ions is very small. The situation should be even worse for higher Z elements.

In the case of the cyclotron this problem is compounded by the high residual pressure near the ion source caused by the relatively high gas flow necessary for its successful operation. The resultant loss due to recombination of the ions being accelerated can become very high. This difficulty can possibly be circumvented by using an external ion source coupled to an axial injector. Another possibility is the use of a linear accelerator to inject an ion with low ϵ across the magnetic field with subsequent transition to high ϵ by means of a stripper foil near the central region. Both of these methods have their own losses and problems and it is by no means certain that great improvements will result by their use.

The ϵ problem poses economic difficulties when one examines the use of the linear accelerator to reach 7 MeV/N. With presently attainable RF gradients it is not feasible to use a single value for ϵ of the ion being accelerated since the machine becomes inordinately long. The usual technique is to start with a low value (0.13 in the case of the Berkeley HILAC) and accelerate the ion to a velocity high enough so that an extremely thin stripper foil can raise ϵ to a value such that the subsequent length of linear accelerator can be of economic length. The HILAC post-stripper tank can now accelerate ions with $\epsilon = 0.25$ but only at about 5% duty cycle. There is an additional loss in the stripping process because of the multiplicity of charge states produced and because of scattering by the foil. These losses can be as much as a factor of 10^2 for heavy ions such as krypton. At the present time we are planning to make a major improvement in the HILAC that will allow it to accelerate up to 10^{11} Kr ions/s average, a thousand times its present capability. This will be done by the substitution of a longer pre-stripper tank equipped with magnetic quadrupole focusing within each drift tube. The design ϵ will be 0.1 or lower and consequently a higher voltage injector is also required. The tentative completion date for this major change is July 1968.

Techniques have been devised involving the use of one or more Van de Graaf accelerators that may be successful in certain respects for the acceleration of very heavy ions to useful energies. The methods usually involve the use of negative ions from the source with a subsequent transition to a higher positive value of ϵ accomplished by passage of the ions through a gaseous medium in the very high positive terminal of a Van de Graaf accelerator (15-20 megavolts). Further acceleration and increase in ϵ is produced by passage of these ions to ground potential through several successive foil strippers. Still higher energies may be achieved by accelerating this beam up to a negative high voltage terminal but then all bombardments must be performed at this high potential. Our study of such accelerators with their many variants has led us to the conclusion that they have marginal utility for our purposes when compared with the other methods available to us.

In 1964 Robert M. Main, Bob H. Smith, and the author conceived the new accelerator system which we call the Omnitron.[6] The ϵ problem which plagues other accelerators is essentially bypassed in this machine since it will accept ions with charge-to-mass ratio as low as 0.05 and still accelerate them to energies as high as 6.5 MeV/N without further stripping. The Omnitron, as presently proposed, consists of two concentric alternating-gradient rings, a rapid-cycling (60 Hz) synchrotron and a dc storage ring, both approximately 120 ft in diameter (see Fig. 5).

There are two possible modes of operation of this system. In the first mode positive ions with ϵ as low as 0.05 are injected from a 3 MV dc accelerator into the synchrotron and accelerated to the desired energy, then transferred to the storage ring from which they are extracted for experiments. The function of the storage ring in this case is to permit long beam spills without slowing down the acceleration process in the synchrotron.

In the second mode of operation, the storage ring is used as part of a double acceleration cycle to produce high-energy heavy ions for biomedical research. As shown in Fig. 6 the cycle begins by the acceleration of beam at a low value of ϵ to the full Bρ of the synchrotron with its subsequent transfer to the storage ring. The ions are held in this ring for 8 ms while the synchrotron guide field decreases to a value appropriate for reinjection of the ions with all or most electrons removed. As the ions are being transferred back to the synchrotron, they are stripped to the higher charge state by passage through a thin foil. They are then reaccelerated in the synchrotron to energies as high as 500 MeV/N. The ion energy output is continuously variable and very well defined in both modes of operation.

We are planning to build two 3 mV injectors so that a great amount of flexibility in operation will be permissible. Most phases of biomedical research do not demand high average beam levels, so that it should be possible to sequentially deliver low-energy beam of one particle to nuclear chemists and high-energy beam of a different particle to biomedical researchers to permit simultaneous use of the accelerator. Figure 7 is a plan view of the system. The low-energy beam gallery is on the left and the high-energy caves are on the right. The AGS rings are in the center and the ancillary equipment is directly above. Fig. 8 is a possible design for the complete building.

Although a synchrotron is basically a very efficient device in that beam once accepted at injection is husbanded carefully all the way through the acceleration and extraction processes to the target, it does suffer from an inherent limitation in its maximum duty factor. Thus with an injection potential of 2.5 MV necessary at an ϵ of 0.05 to achieve 6.5 MeV/N in a single acceleration cycle, the single turn injection time is 28 μs and the duty factor would be 1.6×10^{-3} for a 60 Hz cycling rate. The other basic limitation is that encountered in the early part of the acceleration cycle in the form of a space charge limit which cannot be exceeded without seriously perturbing the betatron oscillations. From a consideration of the aperture that has been proposed and previous experience with working AGS systems this limit is approximately $10^{13}/q$ per second where q is the charge state being used. If the beam available from the ion source exceeds this limit (1 mA) then the duty factor will limit the synchrotron output. In the case of the ultraheavy ions, particularly those with many stable isotopes, the ion source is likely to be the limiting factor. However, by increasing the injection time this problem can be circumvented up to a factor of 30. This can be accomplished by lowering the injection potential and by injecting beam for as many as 10 turns. By this technique saturation of the Omnitron ring can be obtained with as little as 40 μA from the ion source. The low duty factor required of the ion source even under these conditions (5% maximum) will allow its operation at the high arc currents and voltages necessary to produce the high-charge states in the ultraheavy elements (U^{238}, for example, will require a +11 charge).

These general considerations can best be summarized in the following tables prepared by Robert Main which compare the performances of three types of accelerators: (1) a hypothetical linac-injected cyclotron, (2) a hypothetical "super HILAC", and (3) the Omnitron. No comparison is readily applicable for the dc accelerators because of the large number of uncertainties in their performance.

References

1. A. Ghiorso, S.G. Thompson, G.H. Higgins, B.G. Harvey, and G.T. Seaborg, Phys. Rev. 95, 293 (1954).
2. A. Ghiorso, T. Sikkeland, M. Nurmia, Isotopes of Element 102 with Mass 251 to 258. Submitted to Phys. Rev. Letters, February 1967.
3. G. N. Flerov, Yu. Ts. Oganesuan, Yu. V. Lobanov, V.I. Kuznetsov, V.A. Druin, V.P. Perelygin, K.A. Gavrilov, S.P. Tretiakova, V.M. Plotko, Atomnaya Energiya, 17, 310 (1964)
4. W.D. Myers and J.W. Swiatecki, UCRL-11980, May 1965.
   W.D. Myers and J. W. Swiatecki, Nuclear Phys. 81, 1 (1966).
   Other references relating to this field:
   A.G.W. Cameron and R.M. Elkin, Can. Jour. of Phys. 43, 1288 (July 1965).
   Unpublished ORNL report, 1966. Cheuk-Yin Wong, Additional Evidence of Stability of Superheavy Element $^{310}126$ According to the Shell Model.
   A. Sobiczewski, F.A. Gareev, B.N. Kalimkin, Closed Shells for $Z > 82$ and $N > 126$ in a Diffuse Potential Well. Submitted to Physics Letter, August 1966.
5. T. Sikkeland, J.E. Clarkson, D.F. Lebeck, and A. Ghiorso, UCRL 16773, April 1966.
6. A. Ghiorso, R.M. Main, and B.H. Smith. Trans. on Nuc. Sci. 13, 280 (1966).

Table I. Heavy-ion accelerator parameters.

	Linac-Cyclotron	Super Hilac	Omnitron
Injector Megavolts	2.0	2.0	2.5
ϵ	0.06	0.06	0.05
E (MeV/N)	0.120	0.120	0.125
Prestripper	90 ft linac	60 ft linac	-----
Number of drift tubes	228	176	-----
Stripped ϵ (U^{238})	0.17	0.15	-----
E_{max} (MeV/N)	1.75	1.20	-----
RF (Mc/s)	100	100	
Power, RF (MW)	2.7	1.8	
Electric gradient (MV/ft)	0.5	0.5	
Poststripper	125 inch cyclotron	112 ft linac	-----
Number of dees (or drift tubes)	2	138	-----
Spiral angle (deg)	0	---	-----
Acc. Voltage (kV or MV/ft)	75 kV	0.5 MV/ft	-----
B_{max} (kG)	16	---	-----
R_{eff} (in)	57.5	---	-----
RF (Mc/s)	4.0-9.0	100	1.7-33
Power, RF (MW)	0.35	3.4	0.045
Estimated accelerator cost, 1966 ($ million)	11.5	11.0	13.5

Table II. Accelerator performance

	Linac-Cyclotron	Super Hilac	Omnitron
Duty Factor, beam (%)	100	30-100	100
Ion-Source Duty Factor (%)	100	30-100	5 max
Microscopic Duty Factor (%)	20	20	100
Energy Resolution	0.003	0.007	0.0007
Emittance (rad-cm)	10^{-3}	10^{-3}	8×10^{-4}
Variability of Energy	Limited range	Incremental steps, 1-6.5 MeV/N	Continuously variable
Pulsed Beam	With source, beam intensity proportional to width	With source, beam intensity proportional to width	5 µs to dc full intensity
Flexibility[a]	Single energy and particle	Single energy and particle	Complete variation from pulse to pulse possible
System Beam Losses			
Prestripper acceptance	3	3	1.1
Stripping	10	10	--
Poststripper acceptance	7	1	--
Duty factor (U^{238})	1	3	60[b]
Charge exchange	1.6	1	1.1
Extraction	2	1	1.05
Net Loss Factor	670	90	72

[a] Fast transfer or simultaneous delivery of beam to a number of different experimental areas.

[b] For all ions for which the ion-source output is less than 0.1 mA.

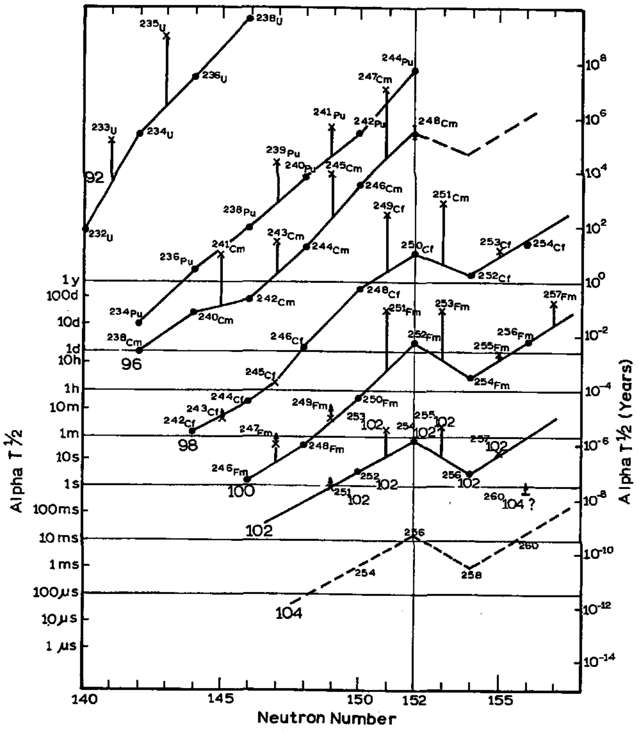

Fig. 1. Alpha half-life vs neutron number for the even Z heavy elements.

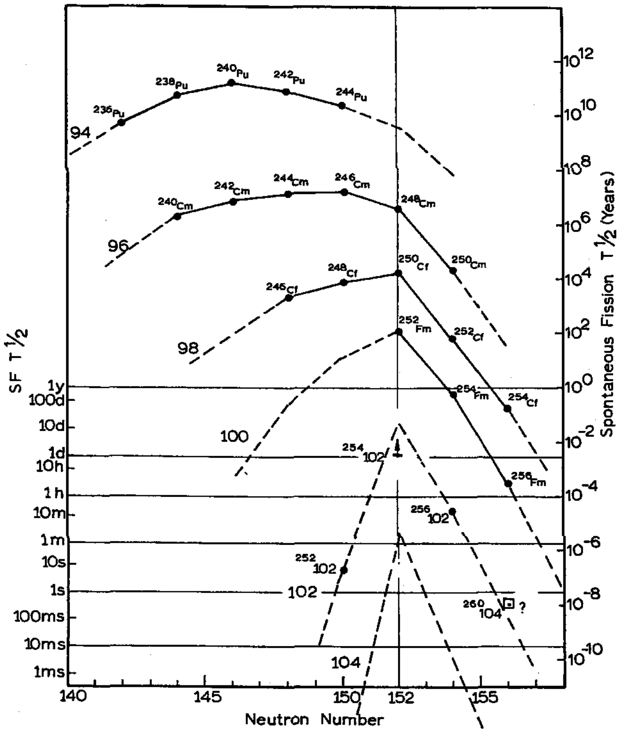

Fig. 2. Spontaneous fission half-life vs neutron number for the even Z elements.

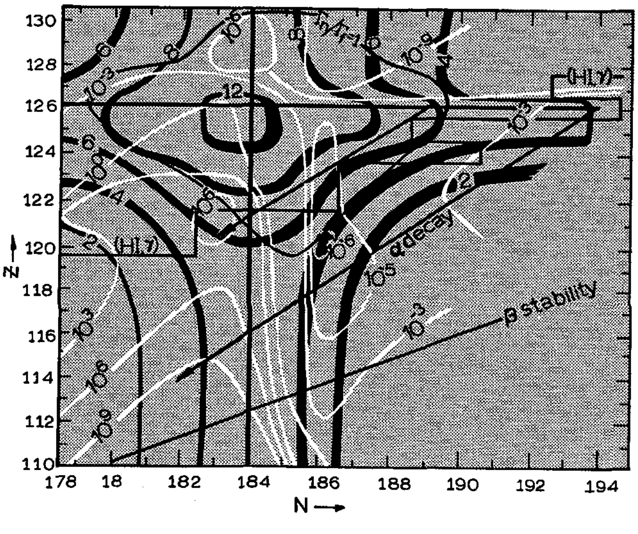

Fig. 3. Hypothetical stability chart for the superheavy elements assuming doubly-closed shells at 126p and 184n. The atomic number Z is the ordinate and the neutron number N is the abscissa. The light contours labeled by figures with exponents are for alpha half-lives in seconds. The heavy contours labeled by figures without exponents are for fission barriers in MeV. The boundary line marked H.I., γ outlines the heaviest nuclides that can be formed by the fusion of stable isotopes with deexcitation only by γ-ray emission.