Heavy Elements Research

Notes based on a series of lectures delivered,

by

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Introduction.

First of all, I should like to say that I feel very privileged to have the opportunity of giving this group of lectures at the Nobel Institute.

I should like to cover a number of topics relating to heavy elements research.

I. Pile and Accelerator Production of Trans-Uranium Nuclides.

First consider the case of pile production. Let us look at an isotope chart and trace the course of the chain of successive neutron capture and radioactive decay processes whereby trans-uranium elements may be produced from parent U^{238} .

U^{238} by radioactive capture of neutrons forms U^{239} , which decays by β^- emission (with half life of 23 min. to Np^{239} , which in turn decays by β^- emission with half life 2.3 days) to the long lived alpha emitter Pu^{239} , which may be recovered chemically from the irradiated uranium.

If the separated Pu^{239} is put back into a pile or if the uranium is left in long enough, one can build up by further neutron capture the heavier Pu

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isotopes, Pu^{240} , Pu^{241} and Pu^{242} . If one has plutonium that has been irradiated long enough to have an appreciable proportion of the beta emitting Pu^{241} (half life = 14 yr) one can remove this Pu and periodically separate chemically the 470 year alpha emitter Am^{241} , which slowly grows in as the beta decay daughter.

The Am^{241} may also be put back into the pile and one produces the two Am^{242} isomers, one having a ~ 100 yr half life and decaying principally by β^- emission, the other having a 16 hr half life and decaying by β^- emission (60 %) to Cm^{242} , by electron capture (20 %) to Pu^{242} , and by isomeric transition (20 %) to the long-lived isomer.

The plutonium produced in an Am^{241} irradiation will consist primarily of the two isotopes Pu^{242} and Pu^{238} (the latter as the daughter of Cm^{242} alpha decay). When such plutonium material is irradiated in the pile, one can produce Am^{243} ($\sim 10^4$ yr α -emitter) and Cm^{244} (19 yr α -emitter), essentially free of the shorter lived α -emitters Am^{241} and Cm^{242} , resp.

The advantages of having weighable amounts of Am^{243} and Cm^{244} lie chiefly in their relatively lower specific alpha activities. Chemical and physical investigations on weighable amounts of Am and Cm in the form of these heavier isotopes may be carried out with more ease and with relatively less health hazard due to radioactivity.

This, then, is the general course that build-up of heavy isotopes from U^{238} by successive neutron capture takes.

Let us proceed now to consideration of the calculation of yields of nuclides formed in chains of successive neutron capture reactions and radioactive decay processes. As a numerical example let us calculate the amount of Pu^{239} obtained following an irradiation of a kilogram of U^{238} at a neutron flux of $\Phi = 3.15 \times 10^{11}$ neutrons $\text{cm}^{-2} \text{sec}^{-1}$ x), for a year

x) Flux of Norwegian-Dutch pile at Kjeller, for example, is given as $10^{12} \text{ cm}^{-2} \text{ sec}^{-1}$.

$(t = 3.17 \times 10^7 \text{ sec}) (\phi = 10^{19} \text{ neutron cm}^{-2}).$

It can be shown that for such a long irradiation as one year it is possible to neglect the relatively short-lived beta-emitting intermediates U^{239} and Np^{239} and to consider the simplified transformation scheme here:

The neutron capture cross section of U^{238} to form U^{239} has been given as 2.80 barns ^{x)} (1 barn = 10^{-24} cm^2). As is well known, the thermal neutron fission cross section of U^{238} is zero. For Pu^{239} the radiative capture cross section (designated hereafter as σ_1^r) is 361 barns and the fission cross section 664 barns, making a total of 1025 barns "destruction" cross section for Pu^{239} (designated σ_2^r).

Designating the number of atoms (or number of moles) of U^{238} at any time, t , as $N_1(t)$, Pu^{239} as $N_2(t)$, the differential equations governing the transformation are

$$\frac{dN_1}{dt} = -N_1 \sigma_1^r \phi$$
$$\frac{dN_2}{dt} = N_1 \sigma_1^c \phi - N_2 \sigma_2^r \phi, \quad (1.1)$$

where $\sigma_1^c = \sigma_1^r = 2.80 \text{ barns}$
and $\sigma_2^r = 1025 \text{ barns}$

The initial conditions are:

$$\left. \begin{aligned} N_1(0) &= N \\ N_2(0) &= 0 \end{aligned} \right\} \text{ For } t = 0 \quad (1.2)$$

It is a simple matter to integrate the first of equations (1.1) to get

$$N_1(t) = N \exp(-\sigma_1^r \phi t) \quad (1.3)$$

which may in turn be substituted into the second of equations (1.1) to give on integration

$$N_2(t) = N \frac{\sigma_1^c}{\sigma_2^r - \sigma_1^r} \frac{\exp(-\sigma_1^r \phi t) - \exp(-\sigma_2^r \phi t)}{\phi} \quad (1.4)$$

x) This and other neutron capture and fission cross sections quoted here are from the declassified compilation of neutron cross sections. AECIL-2040

Substituting numerical values from the given problem into (1.4) one obtains .

$$N_2(1 \text{ yr}) = N \cdot 2.80 \frac{\exp(-2.80 \times 10^{-24} \times 10^{19}) - \exp(-1025 \times 10^{-24} \times 10^{19})}{(1025 - 2.80)}$$

$$= N \frac{2.80}{1022.2} \times 0.01017 = 2.79 \times 10^{-5} N$$

That is to say, about 27.9 atoms of Pu^{239} are obtained for each million atoms of U^{238} initially present. One obtains 28 mg of Pu^{239} per kg of U^{238} . Note that the numerical solution involved a small difference of two exponential terms near unity. One must often carry out such calculations to many decimal places in the large terms in order to obtain a significant final answer.

In a case like this where the total destruction is quite small (i. e. $\sigma_1^T \Phi t \ll 1$) the answer can be more quickly obtained in good approximation by setting $\sigma_1^T \Phi t = \sigma_2^T \Phi t = 0$ and obtaining $N_2(t) = N \sigma_1^C \Phi t$. Numerically in this case $N_2(t) = N \times 2.80 \times 10^{-24} \times 10^{19} = N \times 2.80 \times 10^{-5}$. The error introduced by neglecting destruction is less than 0.4 % here.

The equations (1.1) can easily be extended to a chain of transformations of any length.

$$\begin{aligned} \frac{dN_1}{dt} &= -N_1 \sigma_1^T \Phi \\ \frac{dN_2}{dt} &= N_1 \sigma_1^C \Phi - N_2 \sigma_2^T \Phi \\ &\dots \\ \frac{dN_m}{dt} &= N_{m-1} \sigma_{m-1}^C \Phi - N_m \sigma_m^T \Phi \end{aligned} \quad (1.5)$$

The set of equations is entirely analogous to the equations governing growth and decay in radioactive chains such as the natural radioactive decay chains. In the case of radioactive decay the decay constant, λ ; (where $\lambda = \log_e 2 / \text{half life}$) replaces the product $\sigma \Phi$ in the differential

equations.

Treatments of the general problem have been given previously by several authors ^{x)}.

The general solution (for no two σ_i^r exactly equal) is as follows:

$$N_m(t) = \prod_{1 \leq i \leq m-1} \sigma_i^c \sum_{1 \leq j \leq m} \left[\frac{\exp(-\Phi \sigma_j^r t)}{\prod_{\substack{1 \leq k \leq m \\ k \neq j}} (\sigma_j^r - \sigma_k^r)} \right] \quad (1.6)^{xx})$$

This solution is easily derived by applying the Laplace transform to the differential equations (1.5), converting them into a set of first order simultaneous algebraic equations in the transform. By means of the Laplace transform formalism one can also derive various approximations of the general solution (which for chains of many members may become quite cumbersome) in the form of convergent infinite series. The leading term alone of a power series in t is often useful to estimate yield where destruction can be neglected. That is,

$$N_m(t) \approx \left(\prod_{1 \leq i \leq m-1} \sigma_i^c \right) \frac{(\Phi t)^{m-1}}{(m-1)!} \quad (1.7)$$

if all products

$$\sigma_i^r \Phi t \ll 1.$$

x) Rutherford, Chadwick and Ellis, Radiations from Radioactive Substances. Cambridge University Press, London 1930, p. 15.

Robinson, Journal of Chem. Physics 17, 542 (1949)

Hayden, Reynolds, and Inghram, Phys. Rev. 75, 1500 (1949)

Rasmussen and Barrett, U. C. Radiation Laboratory, Report UCRL 1835, (May 1952)

xx) $\prod$ is the symbol for a general product and the analog of $\sum$, the summation symbol.

A somewhat better approximation may be obtained using the solution for all σ_i^T equal.

$$N_m(t) = \frac{(\Phi t)^{m-1}}{(m-1)!} \exp(-\sigma^T \Phi t) \prod_{1 \leq i \leq m-1} \sigma_i^c \quad (1.8)$$

Finally, it is to be emphasized that pile yield calculations give generally not too good agreement with observed yields. The variation in energy distribution of neutrons between different piles and various other factors make the predictions using "pile neutron" or thermal neutron cross-section uncertain to a large degree.

Miscellaneous Remarks Concerning Pile Irradiations of Heavy Elements.

One will need to design rather safe containers for the pile, particularly where one is irradiating Pu^{239} and materials of comparable or greater specific alpha activity. The container should be of material having a low neutron capture cross section. For instance, aluminum containers sealed by pressure have been used. The first container may be surrounded by another to minimize the possibility of active material leaking to the outside.

One needs to consider the practical engineering problem of heat generation in the sample. In particular, materials with large fission cross sections may generate considerable amounts of heat and necessitate special cooling.

Performing chemical separations on pile irradiated samples often presents many special problems with the high levels of beta and gamma radiation often present. There must be gamma shielding or even remote control manipulators appropriate for the levels of activity being dealt with.

A very valuable tool in this general program of pile production of heavy elements is the mass spectrograph, which enables one to measure

the relative amounts of various isotopes of a given element. For instance, after a long irradiation of uranium one might be most interested in examining the resulting plutonium in the mass spectrograph to determine the relative amounts of masses 239, 240, 241 and 242 present. Knowing the proportion of Pu^{241} present one could calculate at what rate Am^{241} would be growing in.

One also makes considerable use of an alpha pulse analyzer, that is, an ionization chamber with differential pulse height analyzer, since a large number of the heavy nuclides emit alpha particles of characteristic energies at observable rates. One can often analyze isotopic mixtures by alpha pulse analysis. (Pu^{240} and Pu^{239} are a pair which do not lend themselves to relative abundance determination by alpha pulse analysis, since their alpha particle energies are about the same.)

Now, as to predicting the more distant future of heavy isotope production by pile neutron irradiations, one needs particularly to be able to predict or guess the beta stability of undiscovered isotopes. There are a few rules of thumb helpful for this region.

If one examines a plot of the beta stable isotopes vs. atomic number Z and neutron number N , as Fig. 1, with the curved line representing

Fig. 1. Line of Maximum Beta Stability.

the region of maximum beta stability for given Z , one sees that the slope of the line for small Z corresponds to equal increments of Z and N but for the heaviest elements corresponds to addition of two neutrons for each proton. This latter observation, coupled with the fact that beta stability is in large measure dependent on the odd-even character of the nucleus (even Z - even N being relatively most stable, even Z - odd N and odd Z - even N next, and odd Z - odd N least stable), leads to the so-called "Rule of Six", that for the heaviest elements an increase in atomic number of two and mass number of six leads to a nuclide of about the same beta stability. For instance, ${}_{91}\text{Pa}^{231}$ is the only beta-stable isotope of Pa. The rule of six would say that ${}_{93}\text{Np}^{237}$ should be the beta stable Np, which is true. Then, ${}_{95}\text{Am}^{243}$ should be beta stable, but ${}_{95}\text{Am}^{241}$ is also beta stable (by a few kilovolts), showing the rule cannot be taken too literally. The rule would predict ${}_{97}\text{Bk}^{247}$ and ${}_{97}\text{Bk}^{249}$ to be near beta stability. One cannot be certain whether one or both will be stable. The prediction has important bearing on the possible production of Bk by pile irradiations. Cm^{244} is the heaviest isotope reported to have been produced by pile irradiations. The prospects of making much Bk by re-irradiation of Cm^{244} are obviously better if Bk^{247} is the lightest beta stable Bk than if one must produce Cm^{249} to have beta decay to Bk.

For quantitative calculations or estimates of beta decay energies, one makes considerable use of "closed cycle" calculations, employing the principle of conservation of energy over a closed cycle of beta and alpha decays. Figures 2 through 5 are taken from a Lecture Series by Seaborg (Se 52) and show the networks of closed decay energy cycles.

If one adds decay energies around the complete cycle, one must naturally obtain the sum zero.

We might turn now to a consideration of the production of trans-

CLOSED CYCLES - 4n SERIES

Fig. 2

CLOSED CYCLES - 4n + 2 SERIES

Fig. 3

CLOSED CYCLES - 4n+1 SERIES

Fig. 4

[illegible]

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Fig. 5

uranium nuclides by accelerator irradiations. First let us consider some of the special features of the heavy region that will affect bombardment cross sections by the conventional projectiles (deuteron, proton, and alpha particle).

The factors determining reaction cross sections here are generally the same as in lighter mass regions except for competition by the fission reaction, somewhat higher coulombic barriers, and somewhat lower neutron and proton binding energies.

We might first calculate the coulombic barrier for bombardment of U^{238} with protons and with alpha particles. A typical plot of potential energy vs separation distance for the system of a charged particle and a nucleus looks as in Fig. 6.

Fig. 6. Nuclear Potential for Alpha Particles.

Solid line ——— idealized potential.

Dotted line - - - true potential.

The true potential curve might resemble the dotted curve, but it is usually idealized to the solid curve, in which one assumes infinitely short range nuclear forces becoming effective at distances less than the "effective nuclear radius", R . (also called penetration radius or channel radius). Then the potential is assumed a pure coulombic potential at distances greater than R

$$V(r) = \begin{cases} 0 & \text{for } r \leq R \\ zZe^2/r & \text{for } r > R \end{cases}$$

(1.9)

In speaking of "barrier height" we are speaking of a somewhat artificial concept, usually defined as the value of the electrostatic coulomb potential at the effective nuclear radius R

$$B = \frac{z Z e^2}{R} \quad (1.10)$$

A convenient formula for barrier estimation is obtained if one considers

$$R = 1.44 \times 10^{-13} A^{1/3} \text{ cm} \quad (1.11)$$

where A is the mass number.

Since $e^2 = 1.44 \times 10^{-13} \text{ Mev cm}$,

$$B_{(\text{Mev})} = z Z / A^{1/3}$$

There is a good deal of discussion regarding the values one should take for R , even granting its approximate nature. Should R merely be proportional to the cube root of the nuclear mass number, or should it include in addition a constant term to represent the radius of the bombarding particle and the finite range of nuclear forces? That is, what is the value of b in the general expression:

$$R = (r_0 A^{1/3} + b) \times 10^{-13} \text{ cm} \quad (1.12)$$

In the calculations of barrier height here the same assumptions in this regard were made as were made by Weisskopf in his 1947 Los Alamos lecture series (We 47). Namely, Weisskopf assumed an additive constant of $1.48 \times 10^{-13} \text{ cm}$ should be used for alpha particles and none for protons. For the heavier projectiles such as carbon or oxygen ions we assume here that

$$b = r_0 \times a^{1/3}$$

where a is the mass number of the projectile.

That is, we use

$$R = r_0 (A^{1/3} + a^{1/3}) \times 10^{-13} \text{ cm} \quad (1.13)$$

for carbon and heavier ions.

It is not certain what value of r_0 is best to use, so in the table below the barrier heights for protons and alpha particles on U^{238} are given for both $r_0 = 1.4$ and $r_0 = 1.5$.

Table I

Barrier Heights

B in Mev
for U^{238}

Radius coefficient	protons $b = 0$	Alpha Particles $b = 1.48$
$r_0 = 1.4$	16.2	27.6
$r_0 = 1.5$	15.1	25.9

The alpha particle and particularly the proton are light enough particles that there will be some quantum mechanical penetration at energies a few Mev below the barrier height.

The barrier heights in Table I are for the center of mass system. Transformation to the laboratory system will mean an effective increase in barrier height proportional to the ratio of masses of bombarding and bombarded objects. That is, we should add 1.7 % to the alpha barriers and 0.4 % to the proton barriers.

Let us now look at some experimental work on cross sections in the heavy region. Table II gives values from the work of Jungermann (Ju 51) for alpha, fission and deutron, fission cross sections on U^{238} .

Table II.
Experimental Fission Cross Sections.

Particle	Bombarding Energy (Mev)	Fission Cross Section (barns) = (10^{-24} cm ²)
Alpha	18.2 Mev	10^{-3} b
	20.5	10^{-2}
	23.6	10^{-1}
	50	1
	90 - 380	1.6
Deuteron	7.5 Mev	4×10^{-4} b
	9	10^{-2}
	12	10^{-1}
	25	1
	40-190	1.3

For a theoretical discussion of charged particle reactions one may go to Blatt and Weisskopf's recent book (BW 51), page 350 and page 16.

The deuteron cross sections are somewhat more difficult to interpret, since a large part of their reactions involve an Oppenheimer-Phillips mechanism in which only one nucleon of the deuteron interacts with the bombarded nucleus. This mechanism accounts for deuteron reaction cross sections greater particularly at low energies than those derived from simple barrier penetration considerations for the deuteron as a whole.

What can be said about the relative amounts of fission and simple neutron evaporation reactions? The best present information seems to be the work of Tewes and James (TJ 52), who studied radiochemically the relative

amounts of fission products, (p, n) and (p, 3n) products formed by proton bombardment of Th^{232} . For the proton energies ranging from 8 Mev to 21 Mev they found the ratio of fission to the total of the (p, xn) reactions to be about three to one. In accordance with the concept of compound nucleus theory that the de-excitation of an excited nucleus proceeds independently of its mode of formation we would expect this three to one ratio to hold for alpha particle reactions on Th^{232} also. The ratio should increasingly favor fission as we go to compound nuclei of higher atomic number, or more precisely, with greater fissionability parameter, Z^2/A .

A new and promising means for production of tran-plutonium elements is offered by bombardment of uranium with heavier particles than alpha particles. One thus can add more than two units of charge in a single nuclear reaction. Some such transmutation work has been done at the Berkeley 60" cyclotron with carbon ions. It was found when carbon dioxide was introduced into the ion source that an extremely small deflected beam of 120 Mev ions was obtained. It was found later that the internal carbon ion beams were relatively large but were not being extracted into the external beam. Furthermore, there was found a large energy inhomogeneity of the carbon ion beam at a given radius. The best explanation of this curious behavior has been given by the theory of harmonic pre-acceleration. That is, the six-charged carbon ions presumably are not formed in appreciable amounts at the ion source. Instead, doubly charged carbon ions, when formed, are accelerated by the cyclotron in a third harmonic resonant mode. Cyclotron theory gives the result that for a magnetic field and oscillator frequency set to accelerate an ion with a certain charge to mass ratio, the conditions are such that ions with $1/3, 1/5, \dots, 1/(2n+1), \dots$ times the charge to mass ratio will be subjected to harmonic acceleration. The doubly charged carbon ions after some acceleration will possess energies such that collisions

with gas molecules will complete the electron stripping action. The six-charged carbon ion will be normally accelerated then from the point at which the final electron stripping took place. The six-charged carbons will have been formed in a diffuse region and will arrive at the detection probe with a wide spread in energy. Many doubly charged carbon ions are accelerated to the detector also, but they are rather useless for heavy element transmutations, since they have only one third the velocity (i. e., one ninth the energy) of a normal carbon ion. The doubly charged carbons in the Berkeley 60" cyclotron had only 13 Mev and merely contributed a problem due to their heating and burning out foils.

This harmonic pre-acceleration theory has been borne out to a large extent by experiments of Walker and Fremlin (WF 53) with the Birmingham cyclotron. This reference (WF 53) also gives a good review of work done on acceleration of heavy ions. Walker and Fremlin report accelerating several types of heavy ions, many of them not completely stripped of orbital electrons. The Birmingham cyclotron enjoys the advantage over the Berkeley 60" cyclotron that its magnetic field can be increased considerably above the value for the deuteron resonance and thus can accelerate ions of charge-to-mass ratio less than 1:2 (as encountered with incompletely stripped carbon, nitrogen, oxygen ions, etc.). Walker and Fremlin give a plot of the resonance magnetic field vs. the mass-to-charge ratio of the various ions and find the points lie on a straight line, as theory would indicate. Walker and Fremlin's results lend weight to the harmonic pre-acceleration theory, since they find markedly greater beam intensities on the average for six charged ions than for five.

The large cyclotron of the Nobel Institute also has the advantage of being capable of operating with magnetic fields considerably larger than for the deuteron resonance.

While heavy ion beams are of great fundamental interest in the nuclear reaction processes they may induce, they have proved themselves useful tools in production of certain nuclides for study.

Gold foils have commonly been used as monitors for high energy carbon ions, 25 min. and 7 min. astatine alpha emitters being produced by (C^{12} , 4n) and (C^{12} , 6n) reactions, respectively. Alpha activity could not be induced in gold targets by deuterons or alpha particles in the fixed frequency cyclotron. Hence, one need perform no chemical separations but merely count the gold foils in an alpha counter.

There are several regions where the production advantages of heavy ions will be apparent.

In the medium heavy region, such as the lanthanide rare earth region, heavy ion bombardments can produce highly neutron deficient nuclides, free of heavier isotopes. Such bombardments have been applied to study of rare earth alpha emitters.

For the neutron deficient isotopes of Em, Fr, and Ra production cannot be made in the ordinary manner by bombarding target elements of atomic numbers lower by one or two, since macro amounts of these elements are not available. The bombardment with carbon or heavier ions on lead or bismuth thus can be of unique value in isotope production in this region. Momyer, Hyde, and Chiorso (MHC 52) have studied light Em isotopes through carbon ion bombardments of Pb.

Finally, the heavy ion irradiations offer hope as a unique means of production of isotopes in the trans-plutonium region. Chiorso et al (Ch 51) have produced and detected Cf^{246} and Cf^{244} by carbon ion bombardments of uranium.

Let us look in more detail at the prospect of using heavy ion bombardments of uranium.

In estimating cross sections for heavy ion reactions on uranium we shall use the language of Bohr's compound nucleus theory; that is, we assume that the nuclear reactions proceed via a highly excited intermediate nucleus formed by amalgamation of the projectile and the U^{238} nucleus. The de-excitation of this compound nucleus is assumed to proceed independently of its mode of formation. (The compound nucleus has its energy, spin, and parity as constants of motion).

Thus, we estimate first the total reaction cross section for compound nucleus formation in the conventional manner. The potential energy in the U^{238} heavy ion system is assumed as pure coulombic repulsion at separation distances exceeding a certain distance R , at which point shortrange nuclear attractive forces are assumed to set in overcoming the repulsive potential.

It is not clear exactly what values of R should be appropriate, but we shall assume that R can be given by the expression (1.13)

$$R = r_0 (A^{1/3} + a^{1/3})$$

where A is the mass number of the target nucleus and " a " the mass number of the projectile. Barrier calculations will be made with two different values of r_0 , 1.4×10^{-13} cm and 1.5×10^{-13} cm. The barrier height (in the center of mass system) B_{CM} is arbitrarily defined here as the value of the coulombic potential at distance R , that is

$$B_{CM} = \frac{z Z e^2}{R} \quad (1.14)$$

where z and Z are charge numbers of projectile and target and e is the electrostatic unit charge. A certain bombarding energy in the center of mass

system of coordinates corresponds to a somewhat higher energy in the laboratory system, since some energy goes into translational energy of the center of mass. We have the relation

$$B_{\text{lab}} = \frac{(A + a)}{A} B_{\text{CM}} \quad (1.15)$$

Table III gives the calculated values of nuclear potential barriers for bombardment of U^{238} with various ions.

Table III.
Potential Barrier Values for Uranium²³⁸.

Projectile	Barrier (Mev)	for $r_o = 1.4 \times 10^{-13}$ cm	$r_o = 1.5 \times 10^{-13}$ cm
C^{12}	B_{CM}	66.6	62.4
	B_{lab}	70.1	65.5
C^{16}	B_{CM}	87.1	81.4
	B_{lab}	93.1	86.9
O^{18}	B_{CM}	86.0	80.3
	B_{lab}	92.5	86.3

There are undoubtedly corrections which might be added to the simple barrier calculations. Breit, Hull, and Gluckstern (BHG 51) estimate a barrier change due to induced oblate spheroidal distortion of the target nucleus on approach of a projectile. They estimate for O^{16} on U^{238} that the induced

negative quadrupole moment would cause an effective barrier lowering of about 2.9 Mev.

One should instead consider both the quadrupole moment and the effect of a decreased nuclear radius along the line of centers. My own calculations (to second order in a_2 , the spheroidal distortion parameter) show the latter effect predominating over the former and with the O^{16} on U^{238} example give a 6.2 % increase in barrier, or about 5.4 Mev. The effect, while not inconsiderable, is of the same order of magnitude as the uncertainties in choice of nuclear radius. Hence, we shall neglect polarization in the subsequent discussion.

The carbon and oxygen projectiles have much greater mass than protons and hence will behave much more nearly classically. That is, quantum effects, such as, barrier penetration will be of much lower magnitude.

The classical Rutherford cross section formula should give a fair estimate of the total reaction cross section for energies not too near the potential barrier.

$$\sigma = \pi R^2 \left(1 - \frac{B_{CM}}{E_{CM}} \right) = \pi R^2 \left(1 - \frac{B_{lab}}{E_{lab}} \right) \quad (1.16)$$

The formula can be improved to partially account for quantum effects by adding λ , the reduced wave length ($\lambda = h/\text{momentum}$) to R . It can then be shown that

$$\sigma' = \sigma \left[1 + \lambda/R \left(\frac{2E - B}{E - B} \right) \right] \quad (1.17)$$

For example, take a 120 Mev O^{16} ion, $\lambda = 1.04 \times 10^{-14}$ cm.

$$\frac{\lambda}{R} \approx 10^{-2} \quad \frac{2E - B}{E - B} \approx 5$$

hence the cross section would be increased by only 5 % at this energy over the pure classical formula.

Having considered the total nuclear reaction cross section (i. e. the cross section for formation of a compound nucleus), it is now necessary to consider the further course of the nuclear reaction. Several decay processes will be open to the excited nucleus. It may "evaporate" neutrons (and to a lesser extent protons or alpha particles), or it may undergo fission at any time before, between, or after the successive particle evaporation processes, as long as the nucleus has an excitation energy exceeding the fission threshold, a few Mev. Gamma radiation processes are generally much slower than the neutron evaporation or fission processes, hence, gamma radiation will generally occur only when there is left insufficient excitation energy in the nucleus for the other processes.

Let us consider what range of excitation energies the initial compound nuclei might have in heavy ion reactions on U^{238} .

By extrapolating trends of alpha decay energies in the heaviest nuclides on up to the region of as yet undiscovered nuclides Seaborg and Glass (Se 52) have prepared a table of probable relative masses of nuclides to as high as element 102. By use of this table it is a simple matter to calculate the Q -values (enthalpy) of various heavy ion nuclear reactions, as in Table IV.

Table IV.

Estimated Q Values of Some Nuclear Reactions.

Reaction	Q (Mev)	$B_{CM} + Q$ $R = 1.4 \times 10^{-13}$
$U^{238} + C^{12} = 98^{250}$	- 23.9	42.7
$U^{238} + O^{16} = 100^{254}$	- 38.0	49.1
$U^{238} + C^{18} = 100^{256}$	- 38.5	47.5
$U^{238} + N^{14} = 99^{252}$	- 27.1	

The reactions are all highly endothermic. Thus the excitation energy of the compound nucleus will be lower than the energy of the bombarding particle

$$X = E_{CM} + Q \quad (1.18)$$

where X is the excitation energy.

We have already seen that the total cross section will be very small for $E < B$. Thus we see that it is hardly possible to form the compound nucleus with excitation energy less than $B_{CM} + Q$. The third column in Table IV gives this figure for the various reactions. It is obvious then that reactions in which only one, two, or three neutrons are evaporated will occur exceedingly rarely. Experimentally, the $(C, 4n)$ and $(C, 6n)$ reactions are the only ones observed in bombardments of gold and uranium targets in the initial work at Berkeley. Reactions with fewer neutrons evaporated are not seen because excitation of compound nucleus is always great enough to favor evaporation of more neutrons.

Table V gives calculated Q values for various $U^{238} + O^{18} = 100^{256} - x + x n$ reactions, again using the Seaborg-Glass table of masses.

Table V.
Estimated Q -values of $(O^{18}, x n)$ Reaction on U^{238} .

x	Q (Mev)
0	38.5
1	45.0
2	50.5
3	57.1
4	62.9
5	69.9
6	76.2
7	83.2
8	90.0

At the barrier energy of 86 Mev the $(O^{18}, 7n)$ reaction is already energetically possible. More detailed statistical considerations using the nuclear temperature concept to determine the probable kinetic energy carried off by successive evaporated neutrons would lead one to guess that $(O^{18}, 5n)$ is the most probable reaction at the barrier energy. As one proceeds to successively higher bombardment energies the various $(O^{18}, x n)$ reactions will rise to maxima and fall again in the ordinary fashion of $(p, x n)$ or $(\alpha, x n)$ excitation functions.

The $(O, x n)$ reactions will be depressed considerably in magnitude by the competition of the fission reaction. It is difficult to estimate with any

assurance to what extent the fission reaction will predominate. Even for $\text{Th}^{232}(\text{p}, \text{x n})$ reactions with 14-26 Mev Tewes and James (TJ 52) found fission predominating in a ratio of 3:1. The much greater fissionability of nuclei of element 100 will surely mean a much higher ratio than this.

The various considerations discussed in this section are summarized in a semiquantitative way in the excitation function plot of Fig. 7. The absolute magnitudes of the $(\text{O}^{18}, \text{x n})$ cross sections must be regarded as highly uncertain in view of the unknown degree of fission competition (assumed only about 10:1 for the drawing).

In comparing the yields from differently charged projectiles one must remember that the thick target yields will be proportional to an integral

$$\int_0^{E_{\text{max}}} \frac{\sigma(E)}{-\left(\frac{dE}{dx}\right)} dE$$

where $\sigma(E)$ is the reaction cross section as a function of energy, $\left(-\frac{dE}{dx}\right)$ is the rate of loss of energy of the projectile in the target material, and x is the unit of distance through matter.

The much larger values of $\left(-\frac{dE}{dx}\right)$ for carbon or oxygen ions than for protons or alpha particles is a further limitation on the yields of heavy ion reactions.

For the range of energies of interest in nuclear reactions on heavier elements, the ions C^{12} , O^{16} or O^{18} will be completely stripped of orbital electrons in traversal through matter. (The stripping should be essentially complete above 11 Mev for C^{12} . (One makes use of the approximate rule that an orbital electron will be stripped off when the ion velocity through matter exceeds the average velocity of the electron in its orbit.)

Fig. 7. Possible form of excitation functions for reactions of oxygen 18 ions on uranium 238.

The rate of loss of energy of particles of a given velocity traversing matter will be proportional to the square of the charge they bear. For example, deuterons will traverse 730 mg cm^{-2} of lead in going from 25 Mev to 10 Mev, so O^{16} ions, which lose energy 8^2 or 64 times as fast but have 8 times as much energy to lose, will traverse only $1/8$ of this distance (91 mg cm^{-2}) in going between the same velocities (i. e. from 200 Mev to 80 Mev).

For more detailed discussion of the range-energy relationship for heavy ions in matter (including photographic emulsions) one can find excellent reference material in the theses (University of California) of J. I. Miller (Mi 51) and of J. M. Hollander (Ho 51).

II. Systematics of Alpha Decay.

Alpha systematics might be said to consist of a body of semi-empirical correlations involving alpha decay energies and rates and the charge and mass numbers of the various nuclides involved.

Let us first consider some purely empirical correlations to help predict the alpha decay energies and half lives to be expected for undiscovered nuclides of elements 98 and 100. Later we can consider the theoretical bases of the correlations.

If one plots the alpha particle energy on a three dimensional plot against Z and N , it will define a more or less continuous surface. The even-even nuclides show the greatest regularity in decay energies (and particularly α -decay rates) so we choose only them in the following illustration.

The various alpha energy correlations in the literature consist of various two-dimensional representations of this three-dimensional surface.

Figure 8 shows a contour map of the surface, the contour lines connecting equal alpha particle energies. Such a plot has been applied by Glueckauf (Gl 48) and is valuable in giving an over-all picture of the alpha decay energy surface.

In the discussion of the previous section we considered the reactions of oxygen ions on uranium as an illustration of the principles involved. Returning to this example we might ask the question, "What alpha decay energies and half lives might we predict for the new nuclides that might be produced in such heavy ion bombardments?" Some prior estimates of what to expect would greatly facilitate any serious experimental attack on the problem.

Predictions of alpha decay energies of unobserved transitions can be made by interpolation or extrapolation of the alpha energy surface in Fig. 8. One is of course, making the tacit assumption in any smooth extrapolation that no discontinuity in the surface is to be expected. The known surface exhibits a large discontinuity at the neutron shell of 126, but numerous bits of information seem to indicate that we are not approaching very closely another major closed proton or neutron shell near the heaviest nuclides now known. Thus, we may with some confidence make short extrapolations.

For the purpose of extrapolation the contour representation is not as convenient as a plot giving vertical sections through the alpha energy surface. An example of such a representation is the plot of Perlman, Ghiorso, and Seaborg (PGS 50), which plots alpha decay energy vs. mass number, connecting points of the same atomic number. This representation is equivalent to taking vertical cuts through the energy surface in Fig. 8 and viewing them horizontally in the diagonal direction indicated by the arrow.

For predicting decay energies of element 100 nuclides we shall choose to take diagonal cuts at right angles to the arrow in Fig. 8. That is, we shall

Fig. 8. Contour Representation of alpha particle energy surface.
Dots indicate positions of known nuclides, and x's are unknown.

connect nuclides of constant neutron excess (isodiaphères).

Figure 9 shows such a plot, and the extrapolations to three even-even nuclides of element 100 and three of element 98 have been made. Table VI summarizes the results.

If, as we have seen, the alpha decay energy surface is a relatively smooth function of Z and N and since the alpha decay rate of an even-even nucleus is a continuous exponential function of the energy, it follows that a plot of the logarithm of the alpha decay half life vs. Z and N will also define a relatively smooth surface. The two dimensional display of this log half life surface may be made by any of the several ways discussed above for the energy surface. Such plots of log half life vs. mass number at constant Z have been used by Ghiorso (Gh 49).

Such a plot is shown in Fig. 10 where we connect lines of constant neutron excess. The table VI gives the results of the half life extrapolation also.

Table VI.

Alpha Decay Predictions

Z	A	Alpha Particle Energy (Mev)	Alpha Decay Half life
98	242	7.6	20 sec
	248	6.3-6.4	60 day
	250	5.9	20 yr
100	248	7.8	2 sec
	250	7.4	15 min
	252	6.7-7.1	1 day

Fig. 9. Alpha Particle Energy plotted against Mass Number. Nuclides of same neutron excess connected by solid lines. Isotopes connected by dashed lines. o experimental values. x predicted points by extrapolation.

After the simple illustrations of purely empirical application of alpha decay energy and rate correlations we shall proceed to a somewhat more detailed consideration of the principles involved.

First, let us define alpha decay energy in terms of nuclear masses. Since mass and energy are equivalent by the familiar $E = mc^2$ equation, we can write for the alpha decay reaction

the energy balance

$$E_\alpha = 931 (M_Z^A - M_{Z-2}^{A-4} - M_{\text{He}^4}) \quad (\text{in Mev}) \quad (2.2)$$

where the masses are expressed in atomic mass units (masses of neutral atoms).

The alpha particle energy is somewhat less than the alpha decay energy E_α , since by conservation of momentum the decay energy divides itself between alpha particle and recoil nucleus in inverse ratio of their masses

$$E_{\alpha p} = \left(\frac{A-4}{A} \right) E_\alpha \quad (2.3)$$

The concept of "binding energy surfaces" for the system of nuclides is a familiar one, discussed in most texts treating nuclear theory.

One plots, for example against Z and N the mass excess, Δ , where $\Delta = M - A$, (M being the mass of a nuclide in atomic mass units, i. e. ${}^{16}\text{O} = 16.00000$, and A being the mass number.). As Δ is a measure of nuclear instability the energetically more stable nuclides will lie lower than the more unstable. If one includes only even-even nuclides, a fairly smooth surface in three dimensional space is defined. The surface consists of a valley running along the line of greatest beta stability.

Fig. 11. Contour Plot of Mass Excess Surface.

If we next include all nuclides of odd mass number on the plot, a second smooth surface is defined, nearly parallel to and lying just above the even-even surface. (Closer examination points to the odd proton nuclides lying very slightly above the odd neutron members.) Finally, if odd-odd nuclides are included, a third parallel surface lying uppermost is defined.

Neutron or proton binding energy plots vs. N or Z will evidently give sawtoothed curves, since the binding energies involve mass differences of nuclides lying on different surfaces. Note, however, that the alpha decay parent and daughter nuclides are of the same odd or even type, hence will lie on the same surface. It can easily be seen from equation (2.2) for alpha decay energy that

$$E_{\alpha} = 931(\Delta_{Z,A} - \Delta_{Z-2,A-4} - \Delta_{He^4}) \quad (2.4)$$

$$E_{\alpha} \approx 931 \left[2 \left(\frac{\partial \Delta}{\partial Z} + \frac{\partial \Delta}{\partial N} \right) - \Delta_{He^4} \right] \quad (2.5)$$

the partial derivative being evaluated on the surface halfway between the points involved. That is, alpha decay energy is a measure of the slope of the surface, the directional derivative in the direction of equal increments of N and Z .

In regions where the three Δ -surfaces are parallel we shall find that the alpha decay plot vs. N and Z including all nuclides defines a single surface. Such is the justification for including all nuclear types on an alpha decay energy correlation plot such as in Fig. 12.

One may note that there are systematic jogs between odd and even neutron members in certain regions. The odd neutron points generally lie above the trend of even neutron points at neutron numbers just greater than 126, while the reverse is very noticeable in Ra and U. This behavior indicates that the Δ -surfaces are not parallel in these regions, hence that the "pairing energy" of nucleons is undergoing a change with increasing mass number.

It is evident that we can do a fair job in predicting alpha decay energies of odd nucleon types by the same sort of extrapolation or interpolation of the alpha energy surface illustrated earlier. It should not be inferred that the prediction of alpha decay rates can be simply extended to odd nucleon types. Experimentally, irregular and unpredictable hindrance factors act to slow down alpha decay rates of odd nucleon types from the rates of neighboring even-even nuclides of the same energy. One can only predict upper limits on the alpha decay rates of odd-nucleon nuclides.

What about beta decay (including electron capture) energies and rates? Can one apply some sort of systematics to make predictions for undiscovered nuclides?

It is presently possible to make reasonable predictions on beta decay energies for many undiscovered nuclides, but one can do little with regard to predicting decay rates except to set upper limits. (See Figs. 2-5).

The systematization of beta decay energies is not the simple matter of plotting vs. Z and N and defining a surface. Generally only a very few of the beta decay transition of interest in the heavy region have their total decay

Fig. 12 A plot of alpha disintegration energy vs. mass number. From Meinke, Ghiorso, and Seaborg, Phys. Rev. 81, 794 (1951)

energies measured with certainty. The direct measurement of electron capture decay energies has not been done at all except in a few special cases. Fortunately, where we know extensively the alpha decay energies, the measurements of just a few beta decay energies can make possible the calculation of relative masses of many nuclides. One applies the method of closed decay cycle energy balances to calculate the beta decay energies of many transitions. (See, for example, PGS 50 and Figs. 2-5).

One could of course attempt some sort of correlation between beta decay rates and total decay energies. A qualitative correlation must exist, i. e. higher decay energies will lead to shorter half lives. Beta decay rates depend so strongly upon nuclear spin and parity changes that something need be known about the nuclear levels and their spins and parities before attempting to estimate beta decay rates.

One can set a lower limit on the beta decay half life knowing the total decay energy from closed cycle calculations. In no case will the $\log_{10} ft$ value for the beta transition from ground state to ground state be lower than 5, the average value for allowed transitions. (If one takes the prediction of the Mayer-Jensen nuclear shell model that in the heavy region one should expect a parity change in beta decay, then the $\log ft$ values may have 6 as a lower limit, corresponding to first forbidden beta transitions. One should be cautious regarding the shell model parity predictions in the heavy region in view of the presence of states of different parity among the lowest-lying states of Np^{237} .)

We have introduced correlations involving alpha decay rates of even-even nuclides earlier on purely an empirical basis. Let us now look at the theory of alpha decay and see what may be learned from its application to the experimental relationships observed to connect alpha decay rates, decay energies, and atomic numbers.

Most alpha decay rate theories begin with the idealization of the coulombic potential barrier exactly as discussed in the section on heavy ion reaction cross sections. That is, the potential energy for the system of alpha particle and recoil nucleus is considered to be pure coulombic beyond a cut-off distance, R , the "effective nuclear radius for alpha decay", inside which the potential assumes some low value.

Fig. 13. Potential Diagram for Alpha Decay.

The so-called "one body" theories of alpha decay begin with the premise that alpha decay can be treated simply by considering the alpha particle imprisoned within the well created by the potential barrier. There is then a definitely formulated quantum mechanical problem. There exists a small but finite probability for the alpha particle to penetrate the potential barrier and appear in the outside region. The one body problem has been solved in approximate form by various mathematical treatments.

Most simply if we consider that the alpha particle strikes the barrier f times per second, then the alpha decay constant (reciprocal of the mean life) λ can be expressed as

$$\lambda = f \cdot P \quad (2.6)$$

where P is the small probability of barrier penetration. Let us apply for P the simple WKB approximation formula

$$P = \exp\left(-\frac{2}{\hbar} \int_R^{\infty} \sqrt{2M\left(\frac{2Ze^2}{r} - E\right)} dr\right) \quad (2.7)$$

The integration is carried out over the whole region in which V exceeds E .

Substituting $V = 2Ze^2/r$ and integrating one obtains exactly

$$P = \exp [-2g Y(x)] \quad (2.8)$$

where $x = E/B$, $B = \text{barrier height} = 2Ze^2/R$,

$$g = R\sqrt{2MB}/\hbar, \text{ and } Y(x) = x^{-\frac{1}{2}} \arccos x^{\frac{1}{2}} - (1-x)^{\frac{1}{2}}$$

The function $Y(x)$ is plotted by Bethe (Be 37) and is included in a table in the appendix to these notes (Ra 51). By expansion of the $Y(x)$ function in a Taylor series about $x = 0$ one obtains from the first terms an approximate formula which shows roughly the functional dependence of P on various factors.

$$P \approx \exp \left[-\frac{R\sqrt{2MB}}{\hbar} \left(\frac{\pi E}{E^{\frac{1}{2}}} - 4B^{\frac{1}{2}} \right) \right] \quad (2.9)$$

The frequency factor might be taken as just the velocity of the alpha particle divided by twice the nuclear radius. If the velocity within the nucleus in the one body model is roughly equal to the velocity at large separation, we have

$$f = \frac{v}{2R} \approx \frac{10^9}{2 \cdot 10^{-12}} = 5 \cdot 10^{20} \text{ sec}^{-1} \quad (2.10)$$

It is evident from the approximate expression for P that one should obtain a fairly straight plot for a series of even-even alpha emitters of the same element by plotting the log of the half life against $E^{-1/2}$; such is indeed the case.

One of the most elegant solutions of the alpha decay problem on the one-body model is that of Preston (Pr 47), who used a steepest descents approximation for a contour integral representation of the coulomb wave function. This treatment gives nearly the same result as the simple WKB approximation except that there is an energy dependent coefficient before the exponential expression.

With a slight approximation from Preston's original solution (see RTG 53), we can write

$$P_{Pr} = 4 \left(\frac{E}{B-E} \right)^{\frac{1}{2}} \cdot P_{WKB} = 4 \left(\frac{E}{B-E} \right)^{\frac{1}{2}} \exp(-2gY) \quad (2.11)$$

Preston's model brings in a smaller frequency factor than our earlier estimate, since his assumptions give the alpha particle within the nucleus about 0.52 Mev kinetic energy. Hence, with $f = \frac{v}{2R}$ as before, Preston's f will be a factor of 3 or 4 smaller than the estimate in which v is taken equal to the velocity at large separations.

Now the one body model of alpha decay has seemed to many to represent an altogether too naive picture of the nucleus. So called many body variations on alpha decay theory have been proposed, mainly differing from one body theory in introducing smaller frequency factors. Bethe's model (Be 37) proposed

$$F \approx 10^{15} \text{ sec}^{-1}$$

nearly 10^6 lower than our one body estimates. Other many body modifications (RGT 53, Co 50) have proposed frequency factors between the above extremes.

It may at first seem curious that with such a wide range of frequency factors various authors have been able to apply the decay formulas to experimental data to obtain self-consistent sets of results. Note, though, that the usual procedure in alpha decay rate correlation with theory is to leave the effective nuclear radius, R , as an undetermined parameter. Then considering only even-even nuclides one always gets values of R showing a slight dependence on mass number, and plotting R against $A^{1/3}$ one can get quite a good fit with a straight line passing near the origin. The only exceptional even-even nuclides are those with neutron numbers less than or equal to the closed shell number 126. These nuclides, Em^{212} , Po^{210} , Po^{208} decay from 5 to 15 times slower than normal.

Of course, the general magnitudes of R will vary over a 30 % range, depending on the choice of f over the range of 10^6 , but the fits to the data will appear self-consistent, and one does not know enough from other nuclear data

about the magnitude of the effective radius for alpha decay to set the value within 30 %.

An alternative way of correlating even-even alpha decay rates with theory is to assume some radius formula for R and to calculate as undetermined parameter the frequency factor, f . Such a correlation was carried out (Ra 51) using the radius expression $R = (1.43 A^{1/3} + 1.48) \times 10^{-13}$ cm based on an experimental determination of the effective nuclear radius of Th^{232} for alpha particles in nuclear reactions by Jungermann (Ju 50). In this correlation the penetration factor

$$P = \left(\frac{E}{B-E} \right)^{1/2} \exp(-2g\gamma)$$

was used, differing from the Preston treatment value only by the numerical factor 4.

Fig. 14 shows the distribution calculated of $\log_{10} f$ values (designated here as $\log \lambda_0$) for all even-even alpha emitters with well-determined energies and half lives. It is a striking tribute to simple alpha decay theory that such constancy in frequency factor should be found with decay rates ranging over a factor 10^{24} .

The point to be emphasized, though, is that this good self-consistency is not unique to the particular magnitude of nuclear radius chosen. Such could hardly be the case unless the rate of change of penetration factor with energy was nearly independent of the choice of radius. If one takes the approximate expression (2.9)

$$P \approx \exp \left[- \frac{R(2M)^{1/2}}{\hbar} \left(\frac{\pi B}{E^{1/2}} - 4B^{1/2} \right) \right]$$

and evaluates $\frac{\partial \ln P}{\partial E}$ one obtains

$$\frac{\partial \ln P}{\partial E} = \frac{\pi Z \alpha (2Mc^2)^{1/2}}{E^{3/2}} = 0.85 \frac{Z}{E^{3/2}} \quad (2.12)$$

where α is the fine structure constant

$$\alpha = \frac{e^2}{\hbar c} = \frac{1}{137}$$

Fig. 14. Distribution of calculated frequency factors for ground state transitions (alpha) for all known even-even nuclides. Correlation based on radius formula

$$R = (1.43 A^{1/3} + 1.48) 10^{-13} \text{ cm.}$$

Thus, to first order the rate of change of P with energy is independent of R . Hence we cannot determine closely the nuclear radius from alpha decay correlations of this sort.

Fig. 15 plots $\log \lambda_0$ vs. E_α for the correlation in Fig. 14. There is seen to be no systematic trend. Fig. 16 plots $\log \lambda_0$ vs. Z , and we see a definite downward trend.

If, indeed, the frequency factor should be constant as Z increases, agreement could be secured either by a general decrease in the magnitude of the R values or by changing the R dependence on mass number to a lower rate of increase.

It does not seem that alpha decay rate data can provide more than order of magnitude determination of the nuclear radius until our fundamental understanding of the nuclear part of the process is improved. In view of the great success of the penetration factor explanation of alpha decay rate variation it seems that the frequency factor for even-even alpha decay is nearly constant (excluding nuclides with $N \leq 126$). To assume that it is exactly constant and to adjust the R -values accordingly in a rigid manner does not seem justified. With these reservations in mind it might be mentioned that in our article on rare earth alpha emitters (RTG 53) we have tried to include Gd^{148} alpha decay data with heavy element data in various alpha decay rate formulas. With the reservation that the frequency factor in the rare earth region may be nowhere near what the formulas assume, the comparisons seemed to indicate that the most reasonable nuclear radius variation with A is obtained with the formulas employing high frequency factors of the order of 10^{20} sec^{-1} .

It has long been known that many even-even type alpha emitters decayed partly to an excited state and thus showed two alpha particle groups of different discrete energies, so-called "alpha fine structure". Recently alpha

Fig. 15. Trend of calculated frequency factor with Alpha decay energy. (excluding nuclides with 126 or less neutrons.)

Fig. 16. Trend of calculated frequency factor with atomic number. (excluding nuclides with 126 or less neutrons.)

spectroscopic work by Asaro and Perlman (AP 52) at higher resolving power has shown at least two groups for almost all the even-even alpha emitters. The alpha decay rate formulas can be applied separately to each decay group. In general, it is found that decay to the first excited state proceeds with essentially the same frequency factor as to the ground state, but the transitions to higher excited states are often highly forbidden, especially among the heaviest nuclides.

We may define the hindrance factor, η , for excited state transitions in even-even nuclei as the ratio of the frequency factors of excited state and ground state transitions (assuming the same nuclear radius R). Fig. 17 gives a plot of $\log \eta$ vs. A for the various nuclides measured. $\log \eta$ is seen to be very near unity throughout, although the tendency to increase with increasing A seems definite.

Among the odd nucleon number nuclides alpha decay fine structure is almost always present, the number of groups being rather large in several cases. If one calculates the expected alpha decay rates for odd nuclide alpha groups using a rate formula fitted to agree with the even-even ground state alpha transitions, one finds on comparison with the actual decay rates that the odd nuclide alpha groups are often much slower, exhibiting hindrance factors ranging from unity to one thousand.

Our understanding of the basic factors underlying this hindrance is quite limited. There were early attempts to explain this hindrance purely on the basis of the larger potential barrier in the case of non-zero angular momentum of alpha emission, but this effect is now generally felt to be inadequate by itself to explain the very large hindrance factors observed in many cases. Perlman, Ghiorso, and Seaborg (PGS 50) have stressed this point and emphasized that the hindrance should be an intra-nuclear effect associated with the

Fig. 17. Hindrance factor of transition to first excited states for even-even alpha emitters plotted against mass number. Nuclides with same atomic number are connected by solid curves.

probability of assembly of an alpha particle.

There are two absolute selection rules in alpha decay imposed by the fact that total angular momentum and parity are constants of motion and must be conserved during the transition.

If we let ℓ be the angular momentum carried off by the alpha particle, I_i and I_f the spins of initial and final nuclei; and π_i and π_f their parities,

$$I_i + I_f \geq \ell \geq |I_i - I_f| \quad (2.13)$$

and

$$\pi_i \pi_f = (-1)^\ell \quad (2.14)$$

that is, if the nuclear parities are the same, ℓ must be even, and vice versa.

Of great help in achieving understanding of the factors controlling alpha hindrance will be the extensive experimental determination of spins and parities of low-lying nuclear excited states in the heavy region. The problems presented are experimentally difficult because of the very close spacing of levels often observed. Simple nuclear shell model level classifications have not found much application in the region of the heaviest nuclei. It seems hopeful that the Bohr-Mottelson (BM 53) nuclear model combining features of the shell and liquid drop models may provide a helpful theoretical key to level classification in the heavy region.

III. Chemistry of the Heaviest Elements.

(Note: The fourth lecture of the series was devoted to a survey of chemical behavior of the heaviest elements. The lecture was generally a review of points covered in the chemical literature. In these notes the chemistry lecture will be given only in outline form with literature references. The list of references is not intended to be comprehensive, since it consists mainly of references to Berkeley work. Some of the references are review articles giving many other references.)

A. Position of heavy elements in the periodic system. A noble gas electron configuration at 86. 7s, 6d, and 5f orbitals can compete for excess electrons over 86.

Refs. (WB 51) (KM 51)

B. Elements beyond Em chemically may be considered in three groups.

1. Only one oxidation state of importance for aqueous solution chemistry of elements $_{87}\text{Fr}$ through $_{91}\text{Pa}$, namely the 86 electron configuration.

2. Several oxidation states between +3 and +6 of importance for elements $_{92}\text{U}$ through $_{94}\text{Pu}$ (or $_{95}\text{Am}$). Oxidation-reduction chemical procedures convenient.

3. Lanthanide-like elements with only +3 oxidation state of importance, $_{95}\text{Am}$ through $_{98}\text{Cf}$ and probably up through element 103. Rare-earth, fractionation-type chemical separations necessary, notably cation exchange column methods.

Refs. General (SKM 49); separation procedures (Me 49);

Cf (TSGS 50), (STS 50); Bk. (TGS 50), (TCS 50); Cm (WP 51b),

(SS 50); Am (WP 51a); Pu (CW 49), (LS 51); Np (MT 50), (ML 48);

Pa (MGS 51); Fr (H 52); Ion Exchange (KM 50);

C. The debate over electronic configurations of group 2 and 3 elements.
Competition between 5f and 6d orbitals.

Refs. (Se 49), (Da 52).

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Abbreviations

- JACS Journal of the American Chemical Society
UCRL University of California Radiation Laboratory Report
(sent on request from Information Division, U. C. Radiation,
Lab. Berkeley, Calif.)

ERRATA AND ADDENDA TO LECTURE NOTES

Page Equation

5 (1.6) Indices j and k on σ 's in denominator should be transposed.

27 4th line from bottom "ahlfway" should read "halfway"

"Appendix III" should read Appendix I.

References in the Appendix are as follows:

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Appendix III.

Table of $\gamma(y)$ for Alpha Decay Rate Calculations

An exponential factor appears in the alpha particle penetration factor expressions of Bethe⁵⁶ (by the WKB method) and Preston³¹ (derived by a method of steepest descents). This exponential factor is, in the notation of Bethe,⁵⁶

$$P = e^{-2g \gamma(x)} \quad (\text{A } 3.1)$$

where

$$g = (4MZe^2R)^{1/2} / \quad , \text{ independent of decay energy,} \quad (\text{A } 3.2)$$

$$\gamma(x) = \frac{\arccos x^{1/2}}{x^{1/2}} - (1-x)^{1/2} \quad (\text{A } 3.3)$$

$$x = E/B \quad (\text{A } 3.4)$$

E = Total nuclear alpha decay energy

B = The "potential barrier height" = $\frac{2Ze^2}{R}$

M = Reduced mass of alpha particle

R = Effective nuclear penetration radius

e = The electrostatic unit charge

Bethe⁵⁶ presents a graph of the function $\gamma(x)$ vs. x . For precise alpha decay calculations it is desirable to have a table of this function giving γ to greater accuracy than it can be read from the graph. Hence, the following table was prepared.

A change of variable has been made to facilitate calculation. If the substitution

$$x = 1 - y^2 \quad (\text{A } 3.5)$$

is made in (A 3.3), the following expression is obtained:

$$\gamma(y) = \frac{\arcsin y}{(1 - y^2)^{1/2}} - y$$

(A 3.6)

For the compilation of the table values of $\arcsin y$ were taken from the Table of $\arcsin x$ of the New York WPA⁸⁵ and values of $(1 - y^2)^{1/2}$ were taken (as $P_1^1(x)$) from the Tables of Legendre Associated Functions of Mursi.⁸⁶

$\gamma(y)$ is given to five decimal places for the range in y of 0.750 (0.001) 0.946, and values of the first difference are also tabulated.

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TABLE OF $\gamma(y)$

$$y = \sqrt{1 - x}$$

where $x = E/B$

y	$\gamma(y)$	δ_1	y	$\gamma(y)$	δ_1
0.750	0.53215	0.00350	0.790	0.69556	0.00480
1	0.53565	352	1	0.70036	485
2	0.53917	355	2	0.70521	488
3	0.54272	357	3	0.71009	493
4	0.54629	361	4	0.71502	497
5	0.54990	363	5	0.71999	501
6	0.55353	366	6	0.72500	506
7	0.55719	368	7	0.73006	510
8	0.56087	372	8	0.73516	514
9	0.56459	374	9	0.74030	519
0.760	0.56833	377	0.800	0.74549	524
1	0.57210	380	1	0.75073	528
2	0.57590	383	2	0.75601	533
3	0.57973	386	3	0.76134	537
4	0.58359	389	4	0.76671	543
5	0.58748	392	5	0.77214	547
6	0.59140	396	6	0.77761	552
7	0.59536	398	7	0.78313	557
8	0.59934	401	8	0.78870	563
9	0.60335	405	9	0.79433	567
0.770	0.60740	408	0.810	0.80000	572
1	0.61148	411	1	0.80572	578
2	0.61559	414	2	0.81150	584
3	0.61973	418	3	0.81734	588
4	0.62391	421	4	0.82322	594
5	0.62812	424	5	0.82916	600
6	0.63236	428	6	0.83516	605
7	0.63664	431	7	0.84121	611
8	0.64095	435	8	0.84732	617
9	0.64530	438	9	0.85349	623
0.780	0.64968	442	0.820	0.85972	629
1	0.65410	446	1	0.86601	635
2	0.65856	449	2	0.87236	640
3	0.66305	453	3	0.87876	647
4	0.66758	457	4	0.88523	654
5	0.67215	460	5	0.89177	660
6	0.67675	465	6	0.89837	666
7	0.68140	468	7	0.90503	673
8	0.68608	472	8	0.91176	679
9	0.69080	476	9	0.91855	687

y	$\gamma(y)$	δ_1	y	$\gamma(y)$	δ_1
0.830	0.92542	0.00693	0.870	1.27014	0.01084
1	0.93235		1	1.28098	
2	0.93935		2	1.29196	
3	0.94642		3	1.30308	
4	0.95357		4	1.31435	
5	0.96079		5	1.32575	
6	0.96808		6	1.33731	
7	0.97545		7	1.34902	
8	0.98289		8	1.36089	
9	0.99042		9	1.37291	
0.840	0.99802	0.00693	0.880	1.38510	0.01084
1	1.00570		1	1.39745	
2	1.01346		2	1.40997	
3	1.02131		3	1.42266	
4	1.02924		4	1.43553	
5	1.03725		5	1.44858	
6	1.04535		6	1.46181	
7	1.05354		7	1.47523	
8	1.06182		8	1.48885	
9	1.07020		9	1.50266	
0.850	1.07866	0.00693	0.890	1.51667	0.01084
1	1.08722		1	1.53088	
2	1.09587		2	1.54531	
3	1.10463		3	1.55995	
4	1.11348		4	1.57482	
5	1.12243		5	1.58990	
6	1.13148		6	1.60522	
7	1.14064		7	1.62078	
8	1.14991		8	1.63658	
9	1.15929		9	1.65262	
0.860	1.16877	0.01007	0.900	1.66893	0.01084
1	1.17837		1	1.68549	
2	1.18808		2	1.70232	
3	1.19791		3	1.71943	
4	1.20785		4	1.73682	
5	1.21792		5	1.75449	
6	1.22811		6	1.77247	
7	1.23842		7	1.79075	
8	1.24887		8	1.80934	
9	1.25944		9	1.82826	
0.870	1.27014	0.01084	0.910	1.84750	0.01084

y	$\gamma(y)$	δ_1
0.910	1.84750	
1	1.86709	0.01959
2	1.88703	994
3	1.90732	0.02029
4	1.92799	067
5	1.94903	104
6	1.97048	145
7	1.99233	185
8	2.01459	226
9	2.03728	269
0.920	2.06042	314
1	2.08401	359
2	2.10808	407
3	2.13263	455
4	2.15768	505
5	2.18326	558
6	2.20937	611
7	2.23604	667
8	2.26328	724
9	2.29112	784
0.930	2.31958	846
1	2.34867	911
2	2.37843	976
3	2.40887	0.03044
4	2.44003	116
5	2.47193	190
6	2.50459	266
7	2.53806	347
8	2.57236	430
9	2.60753	517
0.940	2.64359	606
1	2.68060	701
2	2.71859	799
3	2.75760	901
4	2.79768	0.04008
5	2.83888	120
6	2.88125	237
		359

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