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SYNTHESIS OF NEW ELEMENTS

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Synthesis of New Elements

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I. AVAILABLE SYNTHETIC METHODS

A. Successive Neutron Capture

The highest thermal neutron flux available to us is about 4×10^{14} neutrons/cm^2/second in the Materials Testing Reactor at Arco, Idaho. Irradiation of hundred milligram amounts of Pu^{239} for about two years will produce, by successive neutron captures and beta decays, about 10^6 alpha disintegrations/minute of the 20-day E^{253} (99^{253}) and about 10^5 alpha disintegrations/minute of the 3.2-hour Fm^{254} (100^{254}). The yield of higher mass numbers is very small. Only a few alpha disintegrations/minute of the 16-hour Fm^{255} have been produced in this way.[1] Fm^{256} was produced by the reaction:[2]

$$E^{255}(n, \gamma)E^{256} \xrightarrow[\text{short}]{\beta^-} Fm^{256} .$$

Unfortunately, Fm^{256} decays with a short half-life (approximately 3 hours) by spontaneous fission. The yield of Fm^{257} would therefore be very small, and in fact none has been observed yet.

Systematics of heavy nuclei (see below) indicate that the lightest isotope of fermium which will decay by beta emission to mendelevium (101) is probably Fm^{259}. The production of this nuclide by the neutron-capture approach is "blocked" by the short spontaneous fission half-life of Fm^{256}, and presumably again at Fm^{258}, which ought to have an even shorter spontaneous fission half-life.

Hence synthesis of element 102 by this method does not look very promising. However, this approach will always be useful for the preparation of long-lived heavy nuclides such as the 2-year Cf^{252} or 1-year isomer of E^{254}, which will

be useful starting materials for other approaches.[3]

B. Thermonuclear Explosions

The very high instantaneous neutron flux produced by the explosion of some thermonuclear devices can cause the rapid successive capture of many neutrons in U^{238} to produce such unlikely nuclides as U^{255}. A chain of successive beta decays then follows, which is finally terminated when the nucleus arrives back on the stability line (in this case Fm^{255} would be obtained).

The nuclides produced in this way resemble rather closely the mixture obtained by method (A), but the isotopic ratios are naturally somewhat different. It was by this method that elements 99 and 100 were first produced and identified.[4]

The method suffers from two obvious disadvantages. First, it is hardly suitable for laboratory application, and the operation of the experiment is quite outside the control of the chemist who merely wishes to produce a new element. Second, the products of the reaction are spread over rather a large area, and it becomes a matter of some difficulty to recover them.

C. Cyclotron Bombardments with Alpha Particles

Cyclotrons such as the Berkeley 60-inch machine can produce beams of alpha particles whose intensities are as high, or even higher, than the neutron intensities available in any reactor. For example, the Berkeley machine can give us a flux of 5×10^{14} 48-Mev alpha particles/cm^2/second. Reaction cross sections in the very heavy element region are small--for example the (a, 2n) peak using Cm^{244} as target occurs at about 20 millibarns.[5] The (a, n) cross section of E^{253} is about 1 millibarn at 40 Mev.

This last reaction was used to produce a few atoms of Mv^{256} for the first time.[6] It should be possible to make several new isotopes of mendelevium in this way, but they will all be neutron deficient, and hence none of them will decay to element 102.

To produce 102 by alpha bombardment would require a target of element 100 (fermium). So far, the longest half-lives known for any of the fermium isotopes are ~16 hours for Fm^{255} and 18 hours for Fm^{252} (made by bombardment of a mixture of Cf^{250} and Cf^{252} with alpha particles.)[7] These half-lives are too short to permit their use as cyclotron targets.

D. Cyclotron Bombardment with Heavy Ions

Cyclotrons at Berkeley, Birmingham and Stockholm have been used to accelerate ions such as C^{12}, N^{14}, Ne^{20}, to energies high enough to produce nuclear reactions in targets such as U^{238}. For example, the Berkeley 60-inch machine accelerated N^{14} to produce $E^{247(?)}$, and the Stockholm and Berkeley machines produced $Fm^{250(?)}$ by the reaction of O^{16} ions on U^{238}. The yields were very low--the Stockholm group only made 20 atoms of Fm^{250}.[8]

Although in principle this is an excellent way of producing new elements, so far none have been first made by the heavy ion method. There are several reasons for this, which are all difficult to overcome.

(1) The beams of accelerated heavy ions are very small--of the order of 0.1 microampere of ions above 100 Mev.

(2) The mechanism of acceleration involves first the production of ions of rather low charge (such as C^{+2}) in the ion source. These ions are then accelerated on a harmonic of the cyclotron RF, and further stripped of electrons by collisions in the D system. Hence the ions which are finally accelerated to high energy are produced over a much larger volume than that of the ion source proper, and a rather diffuse beam with a continuous energy spectrum is obtained.

Very large beams of low-energy low-charge ions fall on the target to produce very severe heating but no nuclear reactions. The problems of target cooling are very serious.

(3) The potential barriers against compound nucleus formation are high. Hence only the upper end of the heavy-ion energy spectrum is available for nuclear reaction. If the target is isolated from the cyclotron vacuum system by even a very thin covering foil, the energy spectrum of the ions is shifted down to lower energies, and a very serious loss in yield occurs. This means that targets must be limited to those nuclides which can safely be placed uncovered in the cyclotron. So far nobody has considered it wise to bombard targets such as Cm^{242} or even Cm^{244}.

(4) Finally, the operation of the cyclotron to produce the maximum possible beams of heavy ions is by no means a simple routine. Conversion from alpha or proton beams requires perhaps half a day. It is therefore convenient to accelerate heavy ions for several consecutive days. Such large blocks of cyclotrol time are not readily available, at least in Berkeley, and therefore heavy ion research programs must be carried out on a "crash" basis two or three times each year.

E. Heavy Ion Linear Accelerator

Both at Yale and Berkeley, linear accelerators are under construction for the sole purpose of accelerating heavy ions. It is expected that they will come into operation towards the end of 1956. They will have several advantages over cyclotrons:

(1) The beams will be monoenergetic. The design energy is 10 Mev per nucleon, i.e., 200 Mev for Ne^{20}.

(2) The beam intensities should be very high--approximately 1 microampere of ions in an area of about 1 cm^2.

(3) The beam comes out into an open, magnetic field-free area where it will be very easy to assemble experimental equipment.

(4) The machine at Berkeley is being built mainly for heavy element research, so that plenty of time should be available.

II. NUCLEAR SYSTEMATICS

An important part of nuclear synthesis is the accurate prediction of the properties of the nuclei to be made. Therefore much attention has been given to this at Berkeley and elsewhere.[9]

A. Alpha and Beta Decay Systematics

The prediction of alpha particle energies and therefore partial alpha half-lives is well understood. The systematic relationship between Z, N and the alpha energy is illustrated in Fig. 1 and is too well known to merit detailed discussion, except to mention the irregularity which occurs in the californium isotopes and probably with fermium too. This has been interpreted as evidence for extra stability--or a minor closed shell--at a neutron number of 152.

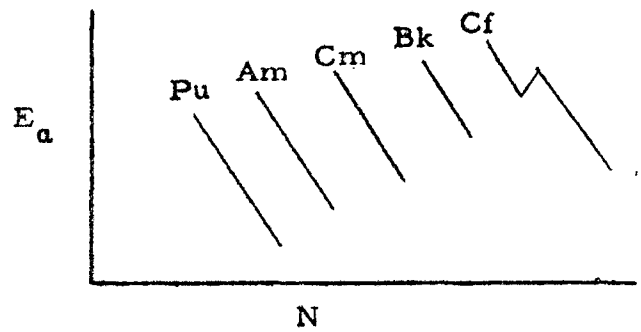

Fig. 1. Systematics of alpha decay energies.

Beta decay energies may be calculated from known or predicted alpha decay energies by means of closed decay cycles of the type shown in Fig. 2.[9]

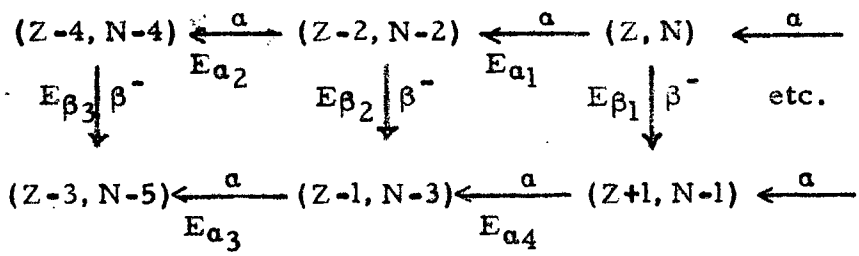

Fig. 2. Closed decay cycles.

The first law of thermodynamics requires that $E_{\alpha_1} + E_{\beta_2}$ shall equal $E_{\alpha_4} + E_{\beta_1}$, also that $E_{\alpha_2} + E_{\beta_3} = E_{\alpha_3} + E_{\beta_2}$. Such cycles may be extended into four two-dimensional networks, one for each of the four radioactive series. If one beta disintegration energy is known in each horizontal row, all the others may be calculated by using measured or predicted alpha disintegrations energies. In many cases, a negative beta disintegration energy would be predicted. This, of course, means that the vertical arrow should be turned around to represent electron capture decay rather than beta emission. If both vertical arrows point towards a given nuclide, it is beta stable.

From the calculated beta disintegration energy, one can make an attempt at the prediction of beta or electron capture half-lives by curves of the Sargent type. The uncertainties are considerable because the degree of forbiddeness of the transition can only be guessed at. However, it is usually possible to set limits which enable experiments to be intelligently planned.

B. Spontaneous Fission Systematics[10]

For the very heaviest even-even nuclides, spontaneous fission may become the chief mode of decay (e.g., Cf^{254}, Fm^{256}). Increasing knowledge has recently revealed a systematic relationship between the neutron number and the fission half-life for even-even nuclides. This is illustrated in Fig. 3.

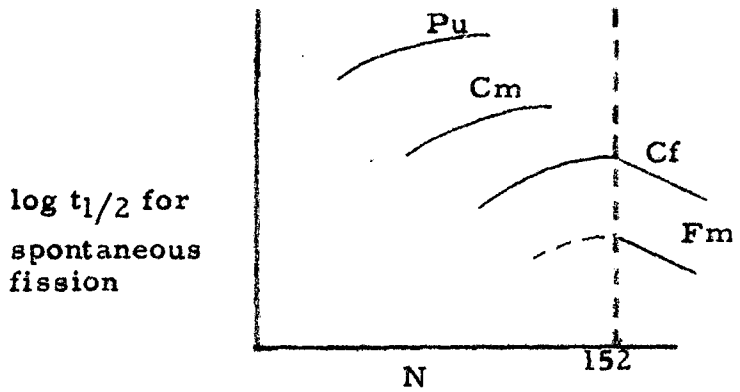

Fig. 3.

Notice that above 152 neutrons the half-lives fall very rapidly. The half-lives for odd nucleon isotopes are longer by about 4 or 5 orders of magnitude than those of the neighboring even-even nuclides. Hence nuclear synthesis in this region will have to depend increasingly on the production of odd isotopes.

C. Cross Sections

It helps to know how much of a new isotope will be produced by a given reaction.

1. Cyclotron bombardments. -- The general features of (α, xn) reaction cross sections are fairly well known. Cross sections probably trend downwards in the heaviest elements. One could safely assume a cross section of about 1 millibarn for an (α, n) reaction, and perhaps 20 millibarns for an (α, 2n) reaction near its peak value.

Cross sections for heavy ion reactions are very small, but no precise information is available because of the energy spread in the cyclotron beams. One does not know the intensity of the ion beam of energy above the Coulomb barrier. There is some indication that reactions such as (O^{16};α, xn) or even (O^{16};3α, 3n) are highly favored.[11] This might be because the barrier is lower for such stripping-type reactions, thus allowing a larger fraction of the beam to become effective.

2. Neutron capture reactions. -- It has been found that there is a rough correlation between the cross section for slow neutron capture and the binding energy of the captured neutron.[12] This is illustrated in Fig. 4. Although approximate, it has been useful in predicting the yields to be expected from successive neutron capture reactions. The neutron binding energies have been calculated by the closed cycle decay method.[9]

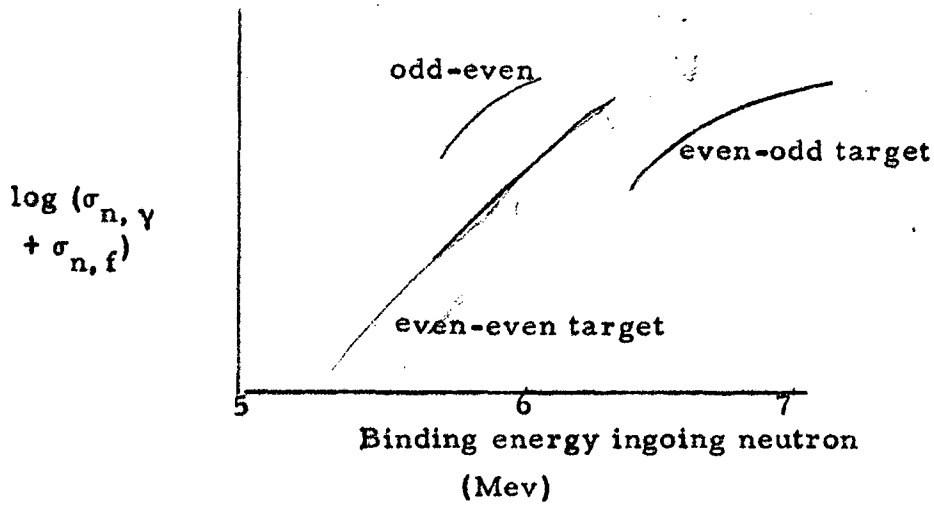

Fig. 4. Prediction of neutron (capture + fission) cross section.

The ratio of fission to (n, γ) cross section is also a steep function of the neutron binding energy. This is illustrated in Fig. 5.[13]

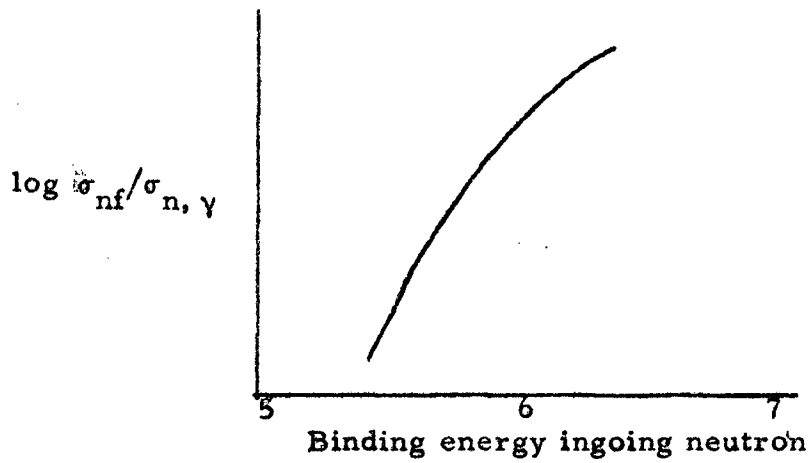

Fig. 5. Ratio of fission to capture cross section.

With measured and predicted cross sections, one can calculate the yield of any nuclide in a chain of successive neutron capture reactions. However, the algebra is very tedious, so H. P. Robinson at Berkeley has built an analog computer which enables calculations to be made with great ease.

D. Chemical Properties

The final isolation of a new element depends on the proper prediction of its chemical properties. In the heavy transuranium region, the actinide hyopthesis of Seaborg, with the associated analogy with the lanthanide series, has been perfectly satisfactory.[14]

All the elements up to mendelevium have been characterized by their elution from cation-exchange columns in the proper sequence. This excellent proof of chemical identity can be completed in about 20 minutes (measured from end of bombardment to start of counting). For nuclides with half-lives much shorter than 5 minutes, more rapid methods may have to be found.

It must be emphasized that there are pitfalls for the unwary. For example, concrete walls exude Rn^{222} which decays finally to Pb^{214}. The Pb^{214} can find its way on to exposed platinum counting discs, especially if they are slightly charged by the presence of high fission product activities. The Pb^{214} elutes from a cation-exchange column as a sharp peak very close to the position of element 100. It subsequently decays to Bi^{214} and Po^{214}, which latter nuclide emits 7.68-Mev alpha particles. With care, one can reduce this background to 1 or 2 events per experiment. Without care, almost 10 times as much contamination may occur.[15]

Obviously every attempt should be made to prove the identity of a new element by adequate chemical tests. This will clearly get more difficult as half-lives get shorter. It is interesting, but not very fruitful, to debate what might be the minimum criterion on which one might claim the discovery of a new element. In the past it was common for a claim to read something like this--"I have observed so and so, which might be attributed to a new element. If this is subsequently confirmed, we claim the discovery and name the element thus and thus." This sort of thing was acceptable when there were no adequate

criteria of chemical identity, but it ought not to be acceptable in 1955.

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