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THE NATURE OF FISSION AND THE CRITICALITY PROCESS

(From Protactinium to Californium and Beyond)

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THE NATURE OF FISSION AND THE CRITICALITY PROCESS
(from Protactinium to Californium and beyond)

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1. INTRODUCTION

The actinide group is composed of 14 elements with atomic numbers 90 - 103; the first three naturally occurring, the other eleven man-made. Although the principal interest in criticality to date has centered on isotopes of the elements U and Pu, a number of isotopes of other elements within the actinide group also are capable of supporting a chain reaction. Calculations or measurements of criticality show extreme variations in critical masses, ranging from gram to kilogram quantities under like environmental conditions. These variations are to be expected, based on simple qualitative considerations and observations concerning nuclear structure and stability.

Neutron fission, a prerequisite for a self-sustaining chain reaction in these elements, depends on the interaction of neutrons with the nucleus. Even for a single given nuclide, extreme variations are likely to occur in the critical mass, subject to a multiplicity of factors affecting the interaction.

Classically, the atom has been likened to a miniature solar system in which electrons orbit about a heavy, dense nucleus composed of neutrons and protons. Quantum theory continues the subdivision, and according to some theories, neutrons and protons may, in turn, be composed of three quarks each - the neutron consisting of two negative quarks and one positive quark; the proton of two positive quarks and a negative quark. To obtain the correct charges for the neutron and proton, the

negative quark would have to carry a charge equal to $1/3$ that of the negative electron, while that of the positive quark would have to be plus $2/3$.

External to the nucleus, the electrons build up in shells, two in the first, eight in the second, eighteen in the third (the equation for the total number of electrons in each shell is $\text{no.} = 2n^2$), etc. The attraction of one atom for another is due to the interaction of the outer electrons and is known as the valence force, wherein lies the basis for the field of chemistry. The source of chemical energy is the rearrangement of the electrons about the nucleus; whereas nuclear energy comes from the rearrangement of nucleons within the nucleus. The closure, or completion of sets of electron shells, or subshells within the principal quantum number, results in atoms that are particularly stable and chemically inactive; under these circumstances, stable compounds with other elements simply do not exist. The "cross section" for chemical reaction could then be considered zero in this case. The inert, or noble gases, He, Ne, Ar, Kr, Xe, Rn, are examples of these types of atoms. These atoms have high ionization potentials; i.e., the energy required to detach an electron has been sharply raised - indicating that the electrons are more tightly bound in these cases.

It is not surprising that the most reactive chemical element is fluorine, an atom short one electron in completing the second shell. Further, due to its high chemical reactivity, fluorine is extremely difficult to separate from its compounds.

The highly successful shell model of the atom serves to explain the sharply increased ionization potentials on closure of shells. A somewhat similar situation prevails in the structure of the nucleus. It is

noted that for certain values of N and Z , there appear discontinuities in the binding energy of the nucleus.⁽¹⁾ These discontinuities occur at the so-called "magic" numbers, 2, 8, 14, 20, 28, 50, 82, 126.⁽²⁾ Nuclei having these numbers of N , Z , or both, are called magic nuclei. The magic numbers appear to be associated with the completion of sets of shells within the nucleus, analogous to the closure, or completion, of sets of electron shells about the nucleus. Closure of a single major nucleon shell results in about 5 MeV of additional binding energy; whereas, the closure of two shells, as in the case of $^{208}_{82}\text{Pb}$ (doubly magic nuclei, $Z = 82$, $N = 126$), gives rise to about 10 MeV of additional binding.⁽³⁾ Neutron capture cross sections are observed to drop sharply on completion of the nuclear shells. The magic nuclei are relatively "inert" for interaction with neutrons, in the same sense that the noble gases (with their completed electron shells) are inert chemically. It also is not surprising, then, to find Xe, which is short one neutron in completing a closed shell of neutrons, to have the highest absorption cross section (2.6×10^6 b) for interaction with the neutron. This is analogous to the case of the electron counterpart, fluorine (short one electron in completing a shell). Consistent with their greater stability, nuclei with magic numbers are anomalously abundant.

It is to be noted, also, that changing from an odd number to an even number of neutrons releases one or two MeV (analogous to the completion of a subshell of electrons), wherein lies the explanation as to why some of the actinides are easily fissionable with slow (low energy) neutrons, but for others, criticality is possible only with unmoderated, or fast, neutrons. With the conclusion of these introductory comments, we now proceed to the sections that follow. The fission and criticality processes,

as such, are reviewed herein on the basis that, "A theory need not be right, provided it is simple." We cite several examples illustrating the complexity of criticality, and then review some of the criticality aspects of actinide elements in general.

2. COMPLEXITY OF CRITICALITY

Criticality, a manifestation of the neutron fission process, is characterized by the attainment of a self-sustaining fission chain reaction. A system is said to be critical if a neutron population, once established, will maintain itself constant, neither increasing or decreasing with time. All factors which influence neutron parameters, or the interaction of neutrons with matter, affect criticality. In order to compute criticality, it is necessary to know the spatial density and neutron cross sections for each kind of atom present in the system. Thus, criticality depends not only on the quantity of fissile material present, but on the size, shape, and material of any containment vessel which may be used, on the nature of any solvents or diluents, and on the presence of any adjacent material that may possibly reflect neutrons back into the fissile material.

Relationship Between Critical Mass and Radius as a Function of Concentration

The following curves (Figure 1), showing the complex variation of critical mass with sphere radius for homogeneous ^{239}Pu -water mixtures, serve to illustrate several effects.⁽⁴⁾ The curves show critical radii of spheres, and critical masses of plutonium contained therein, as a function of water dilution. The upper curve is for bare, homogeneous plutonium-water spheres and the lower one for plutonium-water spheres immersed in water. Striking changes occur in the critical mass as the

ESTIMATED MASS AND RADIUS OF CRITICAL PLUTONIUM-WATER SPHERES

Figure 1

plutonium is diluted with water. Beginning with alpha-phase plutonium metal ($\rho = 19.6 \text{ g/cm}^3$), the critical mass and radius are both seen to increase on dilution with water. The mass passes through a maximum value at an H/Pu ratio of about 4 (Pu density $\sim 5 \text{ g/cm}^3$).

The effect of partially moderating (or slowing down) the fission neutrons causes a significant reduction in the value of η (number of neutrons produced per neutron absorbed in Pu), due to the change in the ratio of the neutron capture and fission cross sections with neutron energy. In addition, the dilution of the metal with water also decreases the density of Pu and increases the neutron leakage. The system must then be made larger to maintain a balance between production and losses of neutrons. On further dilution, moderation by the hydrogen in water becomes increasingly more effective and the probability for fissioning with slow neutrons is enhanced. The effect of adding water is seen to cause a further increase in leakage (and critical size), but the net overall result is a decrease in mass, due to the reduction in Pu density. Finally, on further dilution, or moderation, an optimum condition for production and leakage is obtained, such that the combination results in the smallest critical mass. At this point, the Pu concentration has been reduced to 32 g Pu/l (H/Pu atomic ratio ~ 900). The critical solution volume is about 75 times larger than that for undiluted Pu metal, but the quantity of Pu contained in the sphere is only about 1/10 the metal value. This condition of "optimum moderation" gives a minimum critical mass for the water-reflected sphere that is $\sim 531 \text{ g Pu}$.⁽⁵⁾ Finally, both the critical radius and mass increase once again on further dilution with water, due to excess neutron absorption in the water (principally in the hydrogen). Both become infinite at a plutonium concentration of $7.8 \pm 0.3 \text{ g}$

Pu/H (H/Pu ratio of ~ 3390). At this point, about half the neutrons released in fission are absorbed in the diluent.

Figure 1 shows, also, the effect of neutron reflection. For the sphere immersed in water, some of the neutrons which would otherwise escape are reflected (scattered) back into the sphere, reducing the leakage. The curves show the difference in critical radii brought about by the water reflector.

Triple Point of Criticality

The previous Figure was used to illustrate some of the factors (such as moderation and reflection) that affect the criticality of systems containing a single actinide isotope (for purposes of illustration, ^{239}Pu). The system becomes inherently more complex for mixtures of isotopes. The second most prevalent isotope of plutonium is ^{240}Pu . The effect of the ^{240}Pu isotope on the criticality of ^{239}Pu - ^{240}Pu mixtures is shown in Figure 2.⁽⁶⁾ Calculations indicate that ^{240}Pu could, by itself, be made critical under certain conditions - specifically those under which there would be no moderation by a diluent. The interaction of a thermal neutron with ^{240}Pu results principally in scattering or the formation of ^{241}Pu , since the fission cross section for slow neutrons is negligible. Thus, the effect of ^{240}Pu on the criticality of ^{239}Pu will be largely dependent on the neutron spectrum, which in turn is determined by the concentration and type of diluent present. The curves show the effect of ^{240}Pu on the critical radius, and clearly indicate the existence of triple points of criticality. The effect is more clearly portrayed (schematically) in Figure 3. Note that for a given radius (or fixed volume) there may now be as many as three different critical concentrations! The system would then oscillate between regions that are subcritical and supercritical as

Figure 2

SAME CRITICAL SIZE AT EACH OF THREE CRITICAL
CONCENTRATIONS, a, b, c

Figure 3

Illustration of "Triple Point" of Criticality

a function of fuel concentration. This is brought about by the mere addition or removal of fuel that changes the hydrogen-to-fuel ratio and, consequently, the neutron spectrum.

Density and Effect of Added Scatterers

We shall cite two further examples to illustrate the complexity of criticality before considering the actinide elements more broadly. The variation of critical size and mass with changes in density is of special interest. For a bare system to remain critical, while the density is changed uniformly, all the linear dimensions must be scaled inversely as the density. To maintain the same nonleakage probability or the same number of mean free paths in the system, the dimension and density must be inversely proportional. It follows that the critical mass of an unreflected sphere will vary inversely as the square of the density, $M_c \sim \rho^{-2}$. For infinitely long cylinders, the critical mass per unit length will vary inversely with density, $M_c \sim \rho^{-1}$. In the case of infinite slabs, the mass per unit area, $M_c = \rho^0 = \text{constant}$, and remains unchanged. An unreflected infinite slab which is subcritical remains so, irrespective of the density; criticality in this case could only be achieved by adding more material to the slab so as to increase the mass per unit area. For reflected systems, in which the core and reflector density are varied independently, the variation in the critical mass for finite geometries is given by

$$M_c \propto (\text{core density})^{-m} (\text{reflector density})^{-n},$$

with the provision that $m + n = 2$.⁽⁷⁾

A study has recently been made, however, for the effect of added scatterers (O, C, N) on criticality by means of transport theory calculations utilizing the DTF-IV code.⁽⁸⁾ The study pertained to plutonium

compounds likely to be encountered in fuel fabrication and processing operations in the nuclear industry. It was shown that, in some cases of undermoderated thin slabs with hydrogenous reflectors, the effect of added scatterers in the slab can cause an increase in the critical dimension contrary to the usual expectation that the size should be reduced in such cases. The dominant factor causing the increase in slab thickness was the decrease of neutron leakage into the moderating reflector.

Small Mass Concept⁽⁹⁾

It is now known how, in principle, to obtain a critical configuration with less than an ounce of ^{239}Pu , or with only about one ounce of ^{235}U . These quantities are only about 1/20 of the minimum critical masses prescribed for these nuclides in criticality safety handbooks or safety guides. It should be mentioned that, although these very small quantities could theoretically be made critical, the very specialized conditions required to bring it about would not be encountered by accident. It is possible to cite or reference various other "anomalies" in criticality, but this is outside the scope of the present paper.

3. FISSIONABILITY AND CRITICALITY

(Z^2/A) and Activation Energy

Because the disruptive forces between the protons grow faster than the cohesive forces within the nucleus, a point is reached where the known nuclei terminate with increased Z , due to rapid breakup from spontaneous fission and α decay. Since the deformation, or activation, energy required to induce fission is a function of Z^2/A , this parameter has become known as the fissionability parameter. The limiting condition for nuclear stability, at which point an infinitesimal deformation could cause fission is, $(Z^2/A)_{\text{lim}} = 47.8$.⁽¹⁰⁾

Several years ago, Seaborg developed an empirical relationship for the fission barrier⁽¹¹⁾

$$E_b = (19.0 - 0.36 Z^2/A + \epsilon) \text{ MeV}$$

where

$\epsilon = 0$ for even-even nuclei

$\epsilon = 0.4$ for odd-even and even-odd nuclei

$\epsilon = 0.7$ for odd-odd nuclei

The binding energy of the nucleus may be calculated from the well known formula:

$$\text{Binding Energy} = 931.5 [Z m_H + (A - Z) m_N - M] \text{ MeV}$$

where

$A = Z + N$

M = Exact isotopic weight

m_H = Mass of hydrogen atom

m_N = Mass of neutron

Utilizing these two formulas, the difference between the potential barrier for fission and the energy available on absorption of a neutron (neutron binding energy) can be simply calculated. (Separation energies of the neutron $S(N)$ for all of the elements of interest have now been tabulated by A. H. Wapstra and N. B. Grove.⁽¹²⁾ Simple referral to their tables relieves the necessity of performing the above binding energy calculation.) The activation energy, or energy required to induce fission, was assumed by Seaborg to be 0.9 MeV less than the fission barrier (at the point where time for fission would be comparable to the emission of gamma rays); differences between the neutron binding energy and this activation energy were tabulated for a number of the heavy elements by Vandebosch and Seaborg.⁽¹¹⁾ A negative difference is a measure of the fission thresholds for the various elements.

In Table I, we have included the calculated activation energy and neutron binding energy (and difference) for a number of elements using the notation of Seaborg and have compared these against all of the elements for which critical mass calculations or measurements are known to have been made. (13-17) The Table serves to relate differences and/or correspondence between fissionability, activation energy, and the energy available on absorption of a thermal neutron with data now available on criticality. The table also serves to illustrate the large differences in critical masses that occur between the various isotopes and suggests a distinction be given between nuclides that can be made critical on thermal (as well as fast) neutrons, as opposed to those for which criticality is only possible in the unmoderated state - or with fast neutrons.

Empirical Correlation of k_{∞} vs. Energy Difference Bn-Ea

k_{∞} is defined as the value of the reproduction factor, or criticality factor, in an infinitely large system. A prima facie condition for criticality in any finite system is that k_{∞} shall be > unity. An empirical correlation of k_{∞} vs. energy difference (Bn-Ea) is shown in Figure 4 for even-n nuclides on which computed values of k_{∞} are available. Although there currently is no theoretical explanation, the points group about a straight line which extends from $k_{\infty} = 0.08$ ($^{232}_{90}\text{Th}$ metal) through $k_{\infty} = 2.8$ ($^{244}_{96}\text{Cm}$ metal).

Each of these even-n nuclides falls into our classification of "fissionable" as each has a fission threshold. The probability for fission can be expected to be related to Bn-Ea in a qualitative sense. The value of k_{∞} depends on ν (neutrons per fission), $\frac{\sigma_f}{\sigma_a}$ (ratio of the fission cross section to the absorption cross section) and σ_1 (inelastic

TABLE 1
NEUTRON FISSIONABILITY⁽¹¹⁾ AND CRITICALITY

Nuclide	Z^2/A	E_a (MeV)	B_n (MeV)	$B_n - E_a$ (MeV)	Criticality Aspects		Computed or Measured Critical Masses of Solution Spheres ^(c) Thermal Systems at Optimum Moderation ⁽¹³⁻¹⁷⁾		Computed or Measured Critical Masses of Metal Spheres ^(c) Fast, Unmoderated Systems ⁽¹³⁻¹⁷⁾		
					Slow Neutron Chain Reaction	Fast Neutron Chain Reaction	Bare (kg)	Water Reflected (kg)	Bare (kg)	Water Reflected (kg)	Steel Reflected (kg)
²³² ₉₀ Th	34.91	6.0	4.8	-1.2	No	No	----	----	----	----	----
²³¹ ₉₁ Pa	35.85	5.94	5.6	-0.34	No	Indicated	----	----	----	----	----
²³² ₉₁ Pa	35.49	5.7	6.5	0.8	Questionable	Indicated ^(b)	----	----	----	----	----
²³³ ₉₂ U	36.33	5.1	6.8	1.7	Yes	Yes	1.2	0.59	16.5	7.3	----
²³⁵ ₉₂ U	36.02	5.2	6.5	1.3	Yes	Yes	1.5	0.82	49.0	22.8	----
²³⁸ ₉₂ U	35.56	5.7	4.8	-0.9	No	No	----	----	----	----	----
²³⁷ ₉₃ Np	36.49	5.7	5.5	-0.2	No	Yes ^(a)	----	----	68.6	64.9	43.1
²³⁸ ₉₄ Pu	37.13	5.2	5.6	0.4	No	Yes ^(a)	----	----	7.2	5.6	4.5
²³⁹ ₉₄ Pu	36.97	4.8	6.4	1.6	Yes	Yes	0.905	0.53	10.0	5.42	----
²⁴⁰ ₉₄ Pu	36.82	5.3	5.24	-0.06	No	Yes ^(a)	----	----	158.7	148.4	96.4
²⁴¹ ₉₄ Pu	36.66	5.0	6.3	1.3	Yes ^(a)	Yes ^(a)	----	0.26	----	6.0	----
²⁴¹ ₉₅ Am	37.45	5.4	5.5	0.1	No	Yes ^(a)	----	----	113.5	105.3	71.4
²⁴² ₉₅ Am	37.29	5.1	6.4	1.3	Yes ^(a)	Indicated ^(b)	----	0.023	----	----	----
²⁴³ ₉₅ Am	37.14	5.5	5.4	-0.1	No	Indicated	----	----	----	----	----
²⁴³ ₉₆ Cm	37.93	4.5	6.8	2.3	Yes ^(a)	Indicated ^(b)	----	0.213	----	----	----
²⁴⁴ ₉₆ Cm	37.77	5.0	5.5	0.5	No	Yes ^(a)	----	----	23.2	22.0	14.2
²⁴⁵ ₉₆ Cm	37.62	4.6	6.5	1.9	Yes ^(a)	Indicated ^(b)	----	0.042	----	----	----
²⁴⁷ ₉₆ Cm	37.31	4.7	6.2	1.5	Yes ^(a)	Indicated ^(b)	----	0.159	----	----	----
²⁴⁹ ₉₈ Cf	38.57	4.3	6.6	2.3	Yes ^(a)	Indicated ^(b)	----	0.032	----	----	----
²⁵⁰ ₉₈ Cf	38.42	4.7	5.1	0.4	No	Indicated ^(b)	----	----	----	----	----
²⁵¹ ₉₈ Cf	38.26	4.4	6.2	1.8	Yes ^(a)	Indicated ^(b)	----	0.010	----	----	----
²⁵² ₉₈ Cf	38.11	4.8	4.8	0.0	No	Indicated ^(h)	----	----	----	----	----
²⁵⁴ ₉₉ Es	38.59	4.7	6.0	1.3	Indicated ^(b)	Indicated ^(b)	----	----	----	----	----

E_a = Activation energy for fission (fission time 10^{-14} sec.);
 E_a is 0.9 MeV less than fission barrier.⁽¹¹⁾

B_n = Neutron binding energy for nuclide of mass $A+1$ ⁽¹¹⁾

(a) Computed (see Ref: 14)

(b) No question concerning the possibility of criticality -
but no calculations are known to have been made.

(c) Minimum mass for homogeneous aqueous solutions.

EMPIRICAL CORRELATION OF k_{∞} VS
ENERGY DIFFERENCE $B_n - E_a$
(EVEN- n NUCLIDES)

Figure 4

scattering which can degrade the fission neutrons below the fission threshold). Nonetheless, it is reasonable to expect increased fissioning with higher values of k_{∞} as Bn-Ea increases. Note that ^{238}Pu , once considered somewhat of an oddity (its unreflected critical mass in metal form is less than that of ^{239}Pu), now appears in a rather logical position relative to the other actinide isotopes shown.

An upper limit in the uncertainty for the values of k_{∞} is considered at most no more than ± 0.2 .⁽¹⁸⁾ Some confidence also is manifested in the consistency shown in the k_{∞} values independently computed at BNW and obtained from LASL⁽¹⁸⁾ (private communication with G. E. Hansen). If the empirical correlation is correct, Figure 4 may be used in a qualitative sense to predict the value of k_{∞} for other even-n nuclides. This could be done without any knowledge of the nuclear cross sections of the elements involved, through simple application of the Bn-Ea difference. For example, k_{∞} for $^{250}_{98}\text{Cf}$ (Bn-Ea = 0.4 MeV) should be about 2.50; $^{252}_{98}\text{Cf}$ (Bn-Ea ~ 0) about 1.9, etc.

On examining the critical masses for the even-n nuclides (Table 1), and on referral to the empirical correlation of k_{∞} vs. Bn-Ea, the evidence strongly suggests the achievement of criticality to be possible with a second naturally occurring element that is lighter than uranium, i.e., the protactinium isotope, $^{231}_{91}\text{Pa}$. This isotope would be fissionable with finite critical mass; its k_{∞} might be as high as 1.4.

$^{231}_{91}\text{Pa}$ is somewhat similar to $^{237}_{93}\text{Np}$ in that each is an odd-even nuclide with a slightly negative energy difference: Bn-Ea = -0.34 MeV for ^{231}Pa , and - 0.2 MeV for $^{237}_{93}\text{Np}$. (The computed critical mass for $^{237}_{93}\text{Np}$ is 43 kg, steel reflected.) However, the $^{231}_{91}\text{Pa}$ cross sections required to carry out precise critical mass calculations have not been available.

As is well known, it is only those nuclides with odd numbers of neutrons (those short one neutron in completing a pair) for which criticality is possible with thermal neutrons.

Fissile and Fissionable Nuclei

To describe the criticality properties of the actinide group, it is helpful to refer to "fissile" and "fissionable" nuclei and define the difference.

Fissionable - Those nuclei for which a chain reaction can be set up with the majority of fissions caused by high energy neutrons.

Fissile - Those nuclei for which a chain reaction can be set up with the majority of fissions caused by slow, or thermal, neutrons.

It follows that all fissile nuclei are fissionable but not the converse.

Fissionable Nuclei (Even N Nuclei)

Since fewer studies have been made on the criticality of even N nuclides, as opposed to those with odd N (^{235}U , ^{239}Pu , etc.), the criticality aspects of $^{237}_{93}\text{Np}$, which is typical of the actinides with even N, will be briefly reviewed.⁽¹⁴⁾

The actinides containing even numbers of neutrons in their nucleus characteristically exhibit a rather sharp threshold in fission cross section with little or no probability for subthreshold fission. This phenomenon is illustrated in Figure 5, where the fission to absorption ratio, i.e. the fractional probability for neutron fissioning, is given for $^{237}_{93}\text{Np}$ as a function of neutron energy.

At energies above about 600 keV, neutron absorption results almost entirely in fission, but at lower energies a sharp decrease is observed

^{237}Np FISSION TO ABSORPTION RATIO

Figure 5

to a level where only about 1% or less of the absorptions cause fissions. Achievement of criticality becomes increasingly more difficult as the neutron energy spectrum is degraded into the subthreshold region.

As moderating material is added to a metal-atoms-in-water system, k_{∞} decreases steadily, as shown in Figure 6, from a value of 1.64 for the pure metal at 20.45 kg/l to a value of unity at a ^{237}Np concentration of about 12.7 kg/l. Below 12.7 kg/l, the reproduction factor for an infinite system (k_{∞}) becomes less than unity and criticality is no longer possible. Throughout the concentration range for which criticality can occur, the critical radius for a steel-reflected sphere of metal atoms in water varies uniformly, as shown in Figure 6, from a minimum of 8.0 cm (43 kg) for the pure metal to a value approaching infinity as the minimum critical concentration of 12.7 kg/l is approached.

The effect of energy degradation also becomes evident in the reflector savings of such a system. A good moderating material, such as water, returns neutrons of reduced energy to the core. In the case of $^{237}_{93}\text{Np}$, many of the reflected neutrons are degraded in energy below the effective fission threshold and, hence, those returning neutrons contribute little to the fission process, resulting in a relatively small reflector savings.

The calculations for Np were made from a set of nuclear data compiled and evaluated by B. R. Leonard of Battelle-Northwest.⁽¹⁹⁾ The fission cross sections above 5 keV coincide with those of White, as recommended by Davey.⁽²⁰⁾

4. BEYOND CALIFORNIUM

Increased binding energy at closed, or in the region near closed, shells of nucleons result in increased stability of the nucleus.⁽¹⁾⁽³⁾ Shell closures beyond lead now are predicted at $Z = 114$, and $N = 184$. Due to

CRITICALITY OF HOMOGENEOUS ^{237}Np -WATER SYSTEMS

Figure 6

the additional binding, doubly-closed shell, super heavy "magic" nuclei, if somehow formed, might be relatively stable; whereas, nuclei lying in the region beyond the end of the periodic table prior to reaching this "island of stability" would not exist with any significant half-life. This leads to the prediction of the relatively stable superheavy element $^{298}_{114}\text{X}$, and to others with closed or nearly closed, neutron and proton shells. (3)

J. R. Nix has predicted some of the properties associated with the fission of the hypothetical superheavy nuclei. (3) Some of these properties are included in the following table:

TABLE 2

<u>Fissioning Nucleus</u>	<u>Energy Release Per Fission</u>	<u>Number of Neutrons Per Fission</u>	<u>Lifetime</u>
$^{298}_{114}\text{X}$	317 MeV	10.5	
$^{294}_{110}\text{X}$	290 MeV	10.6	$\sim 10^9$ years *

* Total half-life from decay by spontaneous fission, α decay, β decay, etc.

"Micro" Critical Mass

We may now conjecture as to the possible minimum critical mass of superheavy element(s) X, as yet undiscovered, but theoretically predicted. In making this "qualitative" estimate we have assumed a nucleus one neutron short of the magic number; i.e., a nucleus with an odd number of neutrons but in the region of a closed shell.

^{114}X ; critical radius ~ 3.4 cm
critical volume ~ 0.2
critical mass ~ 0.5 g

* Assuming minimum concentration of ~ 3 g/l in aqueous solution; water-reflected sphere.

The smallest critical mass appearing in Table 1 is only 10 g for $^{251}_{98}\text{Cf}$; a remarkably small value! A nucleus with the properties of "element X" could, however, have a critical mass conceivably only 1/20 that of $^{251}_{98}\text{Cf}$.

5. CONCLUSIONS

Similarities and differences between the shell models for the atom and the nucleus have been used in a simple qualitative manner to help explain in relatively familiar concepts the interaction of neutrons with the actinide elements.

Critical masses range from g to kg quantities depending on the nuclear make-up of the actinide element. In general, criticality is a complex thing - all factors affecting the interaction of the neutron with the nucleus enter in. Several examples were cited in point of illustration, the "triple point" of criticality, apparent anomaly regarding density and effect of added scatterers, the "zero mass concept" in which criticality could, perhaps, be achieved with less than one ounce of ^{239}Pu , etc.

The Table on Slow Neutron Fissionability and Criticality relates differences and/or correspondences between fissionability, (Z^2/A) , activation energy, and the energy available on absorption of a thermal neutron - with the known data now available on criticality. One may classify the actinide isotopes "fissile" or "fissionable" to facilitate discussion of their criticality aspects. From an examination of the material contained in Table I, and the empirical correlation between k_{∞} and Bn-Ea difference shown in Figure 4, it is evident, on the basis of simple qualitative grounds, that it would be possible to achieve criticality with a second naturally occurring element - Protactinium ($^{231}_{91}\text{Pa}$) - that it would be a "fissionable" nuclide, somewhat analogous to $^{237}_{93}\text{Np}$. Comparisons of the kind presented can be used to predict the likelihood of obtaining criticality for

an isotope on which no criticality data exist - but do not provide quantitative information regarding its critical mass. The empirical correlation of k_{∞} vs. energy difference suggests it possible to provide an estimate of k_{∞} for any even-n nuclide without knowledge of either cross section data or ν , on which criticality depends.

Based on the properties of the superheavy "magic" nuclei (as yet undiscovered, but theoretically predicted, and actively sought) an estimate has been provided (on simple qualitative grounds) for critical mass. If the mysterious and illusive element "X" should ever be found, or produced, it might well have a minimum critical mass of the order ~ 0.5 g.

In the field of Nuclear Criticality Safety, one must be prepared to expect the unexpected - to expect and be alert for the "anomalies" therein.

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7. APPENDIX: PROPERTIES OF THE NUCLEUS - FACTS AND SUNDRY

A discussion of the properties of the nucleus involves numbers either so large or so small as to be entirely removed from our everyday realm of experience. The nuclear characteristics are such as to stagger the imagination and, hence, nearly impossible to relate to our conceptual world. Nonetheless, it may be instructive to cite some of these well known properties in hope of obtaining a relative perspective of the nucleus. Nuclear physics is said to be made up from big ideas about exceedingly small things - just how big and how small will become apparent in the examples given.

Density

A single cubic centimeter (about 1/16 cubic inch) of material extracted from the nucleus would weigh $\sim 240,000,000$ tons! Compare this with the density of water (1 g/cc). Obviously, all matter is mostly free space. To be specific, it is space pervaded by powerful fields of electromagnetic force, but fields of force are not of matter or things as we are commonly accustomed.

Surface Tension

The surface tension of water is ~ 75 dyne/cm, which translates into $\sim 3/1000$ oz/cm. The surface tension of nuclear fluid: $\sim 83,000,000$ tons/cm!

Forces Within the Nucleus

Short range (of the order 10^{-13} cm) attractive forces exist between the nucleons (n,n; n,p, and p,p). These forces hold together against the repulsive forces acting between the charged particles (protons) within the nucleus. Coulomb's Law states that like charges repel, and unlike attract, with a force that varies inversely with the square of the distance between, i.e., $F = \frac{q_1 Z_1 q_2 Z_2}{r^2}$. To illustrate the nature of the force, it is calculated

that the repulsion between 2 grams of protons, 30 ft. apart, would be:

111 thousand billion tons!

Although it is common to refer to nuclear energy, it is interesting to note that nuclear fission (with its release of energy) is only made possible because of an atomic long-range force law, Coulomb's Law.

α Decay and Fission

The half-life for ^{239}Pu is $\sim 24,000$ years. ^{239}Pu decays by emission of an α particle; therefore, a single gram of plutonium emits $\sim 2 \times 10^9$ α particles/sec - each with energy ~ 6 MeV. The maximum permissible body burden for plutonium is $\sim 0.65 \times 10^{-6}$ g; ⁽²¹⁾ ($\sim 1.3 \times 10^3$ α disintegrations/sec or ~ 4.7 million/minute). The maximum permissible body burden is that amount of material which, if deposited in the bone, would produce absolutely no observable biological effect during the entire lifetime of the bearer. Yet, it is entirely credible to expect death within a few hours from the biological effects of the fissioning of as little as 0.65×10^{-6} g Pu (or other fissionable material) within the body. This quantity of plutonium is so small as to be scarcely visible to the naked eye without the use of a powerful magnifying glass!

In considering fission and the criticality process, it is apparent the quantities, forces, and effects dealt with, are so far removed from the everyday realm of experience as to render such discussions difficult (if not impossible) in physically meaningful terms.

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