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PLUTONIUM-BERYLLIUM NEUTRON SOURCES, THEIR FABRICATION AND NEUTRON YIELD

By

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The (α,n) nuclear reaction has been utilized for twenty-five years as a source of neutrons. Mechanical mixtures were prepared from an alpha emitter, usually Ra^{226} or Po^{210}, and an element of low atomic number, usually beryllium. Now, however, nuclear reactors produce other alpha-emitting isotopes which can also be used as neutron sources when combined with beryllium.[1] Of the transuranic elements available as products of reactor operation, plutonium is the most abundant. An investigation of the neutron-emitting characteristics of plutonium-beryllium alloys was deemed desirable and such work was started at Los Alamos in 1949.

It was found that plutonium-beryllium alloys make very satisfactory neutron sources for low-flux applications. In particular, the compound $PuBe_{13}$ possesses several advantages over mechanical mixtures of polonium and beryllium or radium and beryllium, although the yield of neutrons per second per cubic centimeter is not as large. The neutron yield and energy spectrum of polonium-beryllium sources vary with the grain sizes of the constituents, as has been pointed out by Stewart.[2] These sources also require frequent time-dependent yield corrections. Disadvantages of radium-beryllium neutron sources include their high cost and their high gamma-ray background. The principal advantage of plutonium-beryllium sources is the stability of the neutron yield with respect to time, which derives from the 24,360-year half life[3] of Pu^{239}. The growth in neutron flux is computed to be only 0.14 per cent in 20 years if suitable plutonium is used. Another important characteristic of $PuBe_{13}$ is that it is the only commonly employed neutron source for which a specific weight of source material has a known and predictable neutron yield.

The metallurgical phase diagram of the plutonium-beryllium binary system has been reported by Konobeevsky[4] and by Schonfeld,[5] and it is characterized by a single compound $PuBe_{13}$ melting at a temperature estimated

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to be about 1950°C. The compound is face-centered cubic and has a measurable range of homogenity.[6] Its density, as calculated from X-ray data, is 4.35 g/cm^3. $PuBe_{13}$ is very brittle; its microhardness exceeds 575 kg/mm^2. It is resistant to oxidation and, unlike many intermetallic compounds of plutonium, does not disintegrate into hazardous powdery material in the laboratory atmosphere.

THE NEUTRON YIELDS

Stewart[2] has determined the neutron spectrum from a $PuBe_{13}$ source and by integration has obtained a total yield of 1.28×10^6 neutrons per second for the source, or 6.1×10^4 neutrons per second per gram of $PuBe_{13}$. Considering the possibilities for error in the method, this value appears to be in reasonably good agreement with an average value of 6.8×10^4 neutrons per second per gram obtained by comparing several specimens of $PuBe_{13}$ with Los Alamos secondary standards. A value of 6.7×10^4 neutrons per second per gram for $PuBe_{13}$ has been reported by Konobeevsky.[4] If, not knowing the isotopic composition, the specific activity of the plutonium used by Runnalls and Boucher[1] is assumed to be 1.4×10^8 disintegrations per minute per milligram, the neutron yield of $PuBe_{13}$ reported by them is calculated to be approximately 6.1×10^4 neutrons per second per gram.

When work on the plutonium-beryllium system was begun at Los Alamos in 1949, calculations were made to predict the neutron yield as a function of alloy composition. The method used was one that had been employed by Bethe[7] in calculating the proton yield of the (α,p) reaction for fluorine as compared to the proton yield of calcium fluoride. Because the form of the plutonium-beryllium phase diagram was completely unknown, and values of the highest possible neutron yields throughout the system were sought, it was assumed in making the calculations that all compositions consisted of a homogeneous single-phase alloy (i.e., the plutonium atoms were considered to be uniformly distributed throughout the beryllium atoms). It is apparent that, with respect to the (α,n) reaction, plutonium acts strongly as a diluent in alloys having a high plutonium content and beryllium similarly dilutes the beryllium-rich compositions, so that the maximum theoretical neutron yield for the hypothetical solid solutions, continuous from pure plutonium to pure beryllium, will occur at some intermediate composition determined as the resultant of two effects: (1) The energy of the alpha particles is dissipated by both plutonium and beryllium atoms in proportion to their numbers and to the stopping powers of the plutonium and beryllium atoms. (2) Alpha particles are supplied for the (α,n) reaction in proportion to the number of plutonium atoms present.

The yield of neutrons per alpha particle from the alloy is inversely proportional to the stopping power of the alloy per beryllium atom, i.e.,

$$\text{neutrons/alpha particle (alloy)} \sim \frac{1}{\frac{N_{Pu} S_{Pu} + N_{Be} S_{Be}}{N_{Be}}} \qquad (1)$$

where S_{Pu} and S_{Be} are the respective stopping powers per atom of plutonium and beryllium and N_{Pu} and N_{Be} are the numbers of plutonium and beryllium atoms. Then, in comparison to pure beryllium

$$\frac{\text{neutrons/alpha particle (alloy)}}{\text{neutrons/alpha particle (pure Be)}} = \frac{S_{Be}}{\frac{N_{Pu}S_{Pu} + N_{Be}S_{Be}}{N_{Be}}} . \qquad (2)$$

Since the number of neutrons/alpha particle (pure Be) is the thick target yield Y for the (α,n) reaction in beryllium,

$$\text{neutrons/alpha particle (alloy)} = Y \cdot \frac{N_{Be}}{N_{Pu}\frac{S_{Pu}}{S_{Be}} + N_{Be}} . \qquad (3)$$

In the computation, S_{Pu}/S_{Be} is assumed to be independent of energy, an assumption which seems to be approximately correct.[8]

The number of alpha particles per second per gram-atom of alloy may be written as

$$\text{alpha particles/sec/gram-atom} = 6.02 \times 10^{23} \lambda \frac{N_{Pu}}{N_{Pu} + N_{Be}} , \qquad (4)$$

where λ is the decay constant for plutonium, i.e., the number of alpha particles per second per plutonium atom.

The product of expressions (3) and (4) is the number of neutrons per second per gram-atom of alloy. This calculation has been made for a series of compositions using the best currently available data for S_{Pu}, S_{Be}, Y and λ. The results are tabulated in Table I and plotted in Fig. 1. The yield of $PuBe_{13}$ is listed in Table I as 18.1×10^5 neutrons per second per 6.02×10^{23} atoms. Conversion of this value to yield per gram of $PuBe_{13}$ gives 7.1×10^4 neutrons per second, to be compared with the best Los Alamos experimental value mentioned above, 6.8×10^4 neutrons per second per gram of $PuBe_{13}$.

If the actual phase diagram of the plutonium-beryllium system represented a continuous series of solid solutions, then the theoretical neutron yields as calculated would be expected. However, the existence of the compound $PuBe_{13}$, and the negligible solid solubility both of plutonium in beryllium and of beryllium in plutonium, give rise to alloys which, except in the case of pure $PuBe_{13}$, consist of crystals of $PuBe_{13}$ distributed throughout a matrix of either plutonium or beryllium. Thus the neutron yield of pure $PuBe_{13}$ should lie on the curve of Fig. 1 and have the value indicated for 92.86 atomic per cent beryllium. But, for all other compositions, the actual neutron yield will be less than that computed for solid solution alloys, and if there were no (α,n) interaction between the

crystals of $PuBe_{13}$ and the matrix phase in which they are contained, the neutron yield per cubic centimeter of alloy would be simply proportional to the volume of $PuBe_{13}$ per unit volume of alloy. On a gram-atomic (instead of unit volume) basis, these yields would lie along the two dashed straight lines in Fig. 1 identified as "rule of mixtures" values.

In Fig. 1 are plotted some experimental points representing the neutron yields of real alloy specimens. It is seen that, in the two-phase alloys consisting of crystals of $PuBe_{13}$ in a matrix of plutonium, the experimental yields, although smaller than the solid-solution values, are always greater than those required by the rule of mixtures. This is because there are beryllium atoms near the surface of the $PuBe_{13}$ crystals that lie within the range of alpha particles originating in plutonium atoms of the matrix. The alpha radiation which passes through the interface from the matrix into $PuBe_{13}$ augments the alpha-particle flux within a zone bordering the interface and thus increases the rate of (α,n) reaction within this portion of the $PuBe_{13}$. Because the surface area to volume ratio depends on crystal size, it follows that, for a given composition of plutonium-beryllium alloy, the smaller the $PuBe_{13}$ crystals contained in the matrix phase, the larger the neutron yield will be. This effect is illustrated in Fig. 1 by the experimental points representing specimens containing different sizes of crystals. An extremely fine grain size of the $PuBe_{13}$ would, of course, approach the condition of uniformly distributed atoms realized ideally in a solid solution or in the crystal structure of pure $PuBe_{13}$. Thus, although higher neutron yields per gram-atom of alloy are obtainable from alloys richer in plutonium than $PuBe_{13}$, only for the exact composition $PuBe_{13}$ is the neutron yield predictable.

In alloys containing more than 92.86 atomic per cent beryllium the $PuBe_{13}$ crystals occur in a matrix of beryllium. Under these circumstances a much smaller contribution to neutron yield additional to the rule-of-mixtures value results from a flow of alpha particles across the interface between the $PuBe_{13}$ and the beryllium matrix. In this case, alpha particles from plutonium atoms within the $PuBe_{13}$, but near the surface of the crystals, react with beryllium atoms in the matrix, as well as with those within the compound. Although not shown in Fig. 1, experimental values for the neutron yields of these alloys were found to lie in the narrow region between the straight line and the curve at the extreme right of Fig. 1.

Runnalls and Boucher[1] have demonstrated nicely the dependence of neutron yield on the form and aggregational state of the component elements. In an investigation of beryllium-rich alloys of plutonium they observed, among other similar effects, a marked increase in neutron yield when the alloys melted.

Because plutonium is a product of the nuclear reactor, its isotopic composition is a function of reactor characteristics and operation. The stability of the neutron yield of a plutonium-beryllium source depends on the 24,360-year half life of Pu^{239}. Other isotopes present are Pu^{238},

Pu^{240}, and Pu^{241}. The amount of Pu^{238} with its 89.6-year half life in currently available plutonium is relatively small and the larger amounts of Pu^{240} have a 6580-year half life. The effect of these isotopes on the rate of emission of neutrons is not significant for periods of ten to twenty years. If, however, an appreciable amount of Pu^{241} is present, then the alpha-active daughter Am^{241} with its 12.9-year half life alters the number of alpha particles per second per gram-atom and the virtue of a neutron source of constant yield is lost.

Coon[9] has calculated that the growth in rate of emission from a plutonium-beryllium source is related to the Pu^{241} content in the following manner:

$$\frac{Q_t}{Q_o} = 1 + k \left[1 - \exp\left(-t/18.6\right) \right] ,$$

where

Q_t = the neutron emission rate at the time t years,
18.6 = the mean life of Pu^{241} in years,
t = the time in years from the start of Am^{241} accumulation due to beta decay of Pu^{241},
and Q_o = the neutron emission rate in the absence of any Am^{241}.

The quantity k is obtained from the following expression:

$$k = \frac{1.27\, a^{Pu^{241}}/T^{Am^{241}}}{a^{Pu^{239}}/T^{Pu^{239}} + a^{Pu^{240}}/T^{Pu^{240}} + 1.27\,(a^{Pu^{238}}/T^{Pu^{238}})} ,$$

where

a = the relative abundance of the isotope,
and T = the half life of the isotope.

The numerical factor 1.27 appearing in this expression for k is the ratio of the number of neutrons produced by 5.48 Mev alpha particles (Am^{241} and Pu^{238}) and by 5.14 Mev alpha particles (Pu^{239} and Pu^{240}). This numerical value is taken from the experimental work of Runnalls and Boucher.[1]

As a numerical example, the growth in the rate of neutron emission is 2.6 per cent in 20 years from a plutonium-beryllium source prepared from plutonium containing 0.06 per cent Pu^{241}. The growth is only 0.04 per cent in 20 years from a similar source prepared from plutonium containing 0.003 per cent Pu^{241}. Thus it is clear that the most useful neutron sources to be obtained from plutonium and beryllium have exactly the composition $PuBe_{13}$ and are fabricated from plutonium containing a minimum amount of Pu^{241}.

FABRICATION OF THE SOURCES

Like all alloys of plutonium, those of plutonium and beryllium are prepared in suitably equipped gloveboxes in order to minimize the hazards of handling the plutonium. The first plutonium-beryllium alloys were prepared at Los Alamos in 1950 by F. W. Schonfeld, C. R. Tipton, and R. D. Moeller. A satisfactory method for preparing them is to weigh appropriate amounts of the two metals into a beryllium oxide crucible. It is important to load the heavy plutonium metal on top of the lighter beryllium metal. Because the size of the melts is kept small for health physics reasons, it is helpful to load a single piece of each metal in order to obtain good alloying. If several small pieces are loaded, some may hang onto the crucible wall and not enter the melt. The crucible is heated by means of a tantalum susceptor in an induction furnace containing an argon atmosphere. At compositions corresponding to $PuBe_{13}$ the two elements react vigorously as the temperature approaches 1150°C, and the heat of this reaction suddenly carries the temperature of the small mass to approximately 1400°C. This exothermic reaction yields a friable mass having the character of coke. If the mass is further heated to about 2000°C it coalesces. Upon cooling, a hard, brittle ingot of $PuBe_{13}$ is obtained which possesses evidence of considerable solidification shrinkage. Runnalls and Boucher[1] have reported another method of preparation, namely, the reduction of plutonium trifluoride by powdered beryllium. After the reduction, beryllium trifluoride is distilled off leaving a fluoride-free alloy of plutonium and beryllium.

The alloys are encapsulated in order to permit their being handled in the laboratory without danger of spreading radioactive contamination. Capsules suitable for containing $PuBe_{13}$ should meet the following requirements:

1. They must be rugged in order to minimize the possibility of breaking a container.

2. They must be easily loaded and permit rapid sealing in order to minimize neutron exposure to personnel preparing the sources.

3. The seal must be tight in order to preclude the possibility of spreading radioactive material.

4. Magnetic containers are desirable, as they lend themselves to remote handling by magnetic methods.

Three styles of containers which have been evolved at Los Alamos are illustrated in Fig. 2. The one-inch cylindrical container was designed for a source strength of 10^6 neutrons per second, the larger spherical container for 4.5×10^5 neutrons per second, and the smaller spherical container for 6×10^4 neutrons per second. Nickel has proven to be a satisfactory material from which to machine these capsules.

Loading and sealing the capsules is done in gloveboxes. The cylindrical container (Fig. 2a) is loaded with crushed $PuBe_{13}$. Lumps of the compound, either the coke-like material or dense material produced by

melting, are placed in the container. The lumps are simultaneously crushed and packed to a bulk density of approximately 3.7 g/cm^3 by ramming them with a suitable tool. The spherical containers (Figs. 2b and 2c) are loaded with a lump of material that has been melted and solidified in a beryllium oxide crucible. Frequently, in breaking the crucible away from the compound, the lump of compound is broken. This may make it difficult to fit the material into the container. Even if the lump is a single piece, the most compact source suggested by the X-ray density is not obtained because of pipe formed in the ingot on solidification.

Capsules are sealed by induction brazing, using a preplaced hard solder ring. A solder containing 56 per cent silver, 22 per cent copper, 17 per cent zinc, and 5 per cent tin (American Platinum Works Silvaloy No. 355) and a paste-type flux containing fluorides and borates (Handy and Harmon Handyflux) have been found to give satisfactory results. The joint and solder are coated with a minimum amount of flux, the solder ring is positioned, and the flux is permitted to dry before the capsule is placed in the contaminated glovebox. After the capsule is loaded, it is placed in a soldering jig. For the smallest source the soldering jig is also used to hold the capsule during loading. Heat for soldering is applied by means of a single-turn coil connected to a RF transformer. After soldering, traces of oxidation and flux are removed from the capsule by pickling it in a hot solution of hydrochloric acid and cupric chloride. It is then rinsed in hot water.

Before each capsule is considered to be satisfactory, it must pass a leak test. This test is conducted by placing the capsule in a small pressure vessel in which a helium atmosphere is raised to a pressure of 200 psi. After 30 minutes the pressure is released, and the capsule is dropped into ethanol or a similar liquid having low surface tension. Leaks are indicated by helium bubbles streaming from the capsule. Containers which leak may be resoldered, or they may be opened and the material recanned.

Because all of the canning operations have taken place in a group of contaminated gloveboxes, the exterior of the capsule is contaminated and must be cleaned. This is done best by scrubbing the capsule to remove loose material from the surface and then vapor plating it in an atmosphere of nickel carbonyl to form a coating 5 mils thick.

The final step in the preparation of these sources is to have them calibrated in a graphite column using the technique described by Graves and Froman.[10]

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TABLE I

Calculation of the Theoretical Neutron Yields of Plutonium-Beryllium Alloys

Atom Fraction Beryllium	$N_{Pu}\frac{S_{Pu}}{S_{Be}}$	$\frac{N_{Be}S_{Be}}{N_{Pu}S_{Pu}+N_{Be}S_{Be}}$	Neutrons per Alpha Particle	Alpha Particles per sec per 6.02×10^{23} atoms	Neutrons per sec per 6.02×10^{23} atoms
0.00	5.88	0.0000	0.00	54.2×10^{10}	0.0
0.10	5.30	0.0185	1.26×10^{-6}	48.8 "	6.2×10^{5}
0.20	4.71	0.0407	2.77 "	43.4 "	12.0 "
0.30	4.12	0.0677	4.60 "	37.9 "	17.4 "
0.40	3.53	0.1018	6.92 "	32.5 "	22.5 "
0.50	2.94	0.1454	9.88 "	27.1 "	26.8 "
0.60	2.35	0.2063	14.0 "	21.7 "	30.4 "
0.70	1.77	0.2835	19.3 "	16.3 "	31.5 "
0.80	1.18	0.404	27.5 "	10.84 "	29.8 "
0.90	0.59	0.604	41.1 "	5.42 "	22.3 "
0.9286*	0.42	0.689	46.8 "	3.87 "	18.1 "
1.00	0.00	1.000	68.0 "	0.00 "	0.0 "

*$PuBe_{13}$

Notes on the experimental data used in the calculations:

1) The mean energy for alpha particles from Pu^{239} is 5.14 Mev.[11]

2) The experimental stopping power of plutonium is not available. The stopping power of lead for alpha particles is used as an approximation. The mass stopping power of lead for 5.14 Mev alpha particles[12] is 0.225 Mev/mg/cm^2.

3) The experimental stopping power of beryllium for alpha particles is not available. The stopping power for protons is converted to the stopping power for alpha particles by the relation

$S_\alpha = 4\ S_p$ at the same velocity; i.e., at one-fourth the energy.

The mass stopping power of beryllium for 1.25 Mev protons[12] is 0.220 Mev/mg/cm^2. Thus, the mass stopping power of beryllium for 5 Mev alpha particles is computed to be 0.88 Mev/mg/cm^2.

4) The mass stopping powers are given in footnotes (2) and (3). However, atomic stopping powers are required for the ratio S_{Pu}/S_{Be}. The atomic stopping power is related to the mass stopping power by the relation[13]

$$S_a = \frac{S_m A}{N}$$

where A is the atomic weight and N is Avagadro's number. The ratio of the atomic stopping powers is, therefore,

$$\frac{S_{Pu}}{S_{Be}} = \frac{(0.225)\ (207)}{(0.88)\ (9)} = 5.88$$

5) The thick target yield of beryllium for 5.14 Mev alpha particles is 68 neutrons per 10^6 alpha particles.[14]

6) The decay constant of plutonium is computed from the 24,360-year half life[3] by the relation

$$\lambda = \frac{0.6931}{T}$$

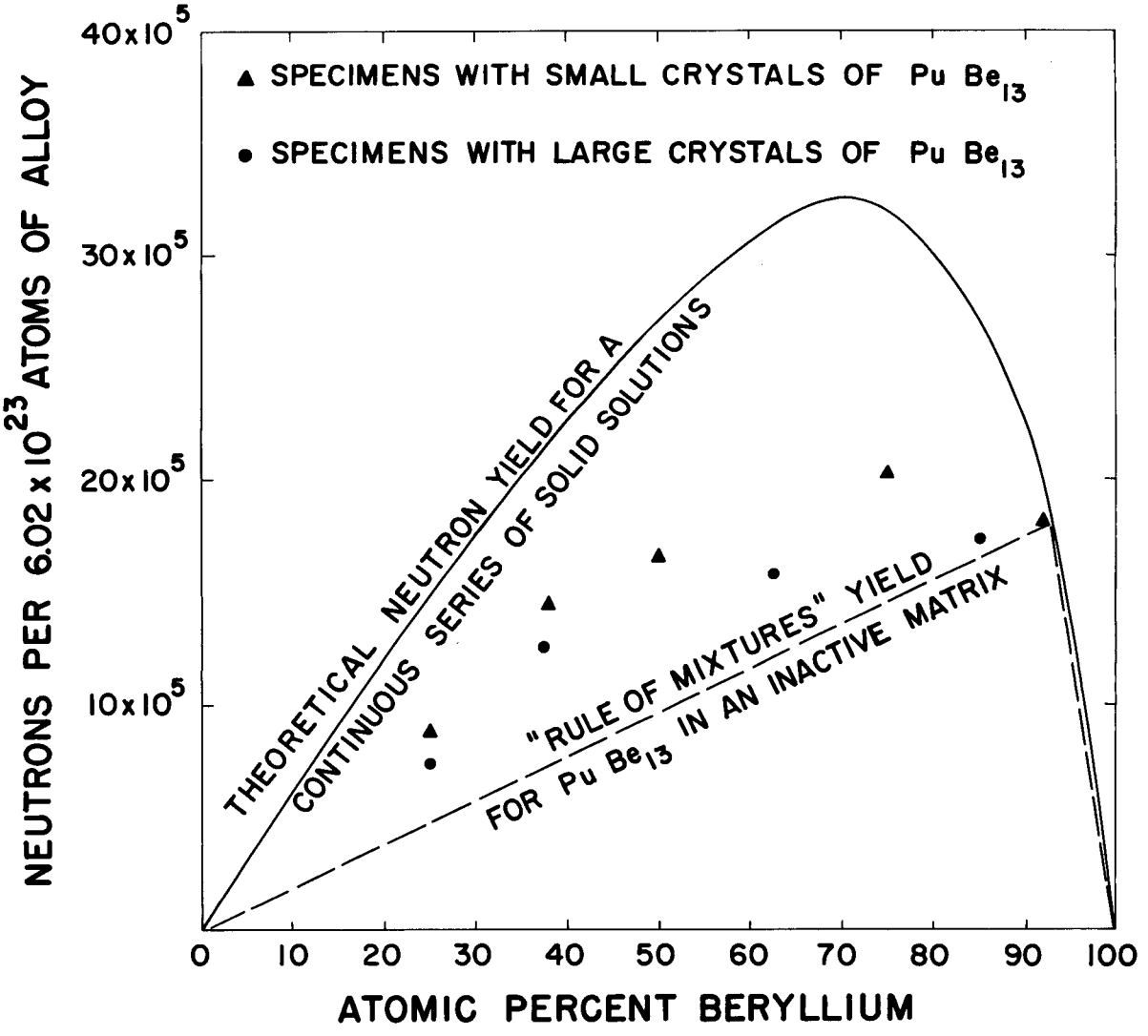
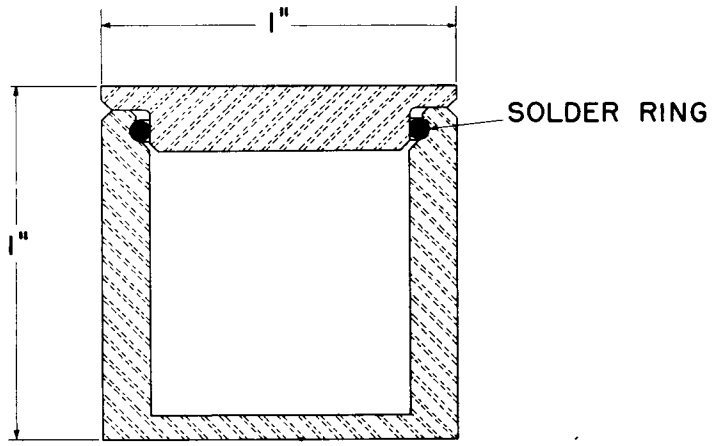
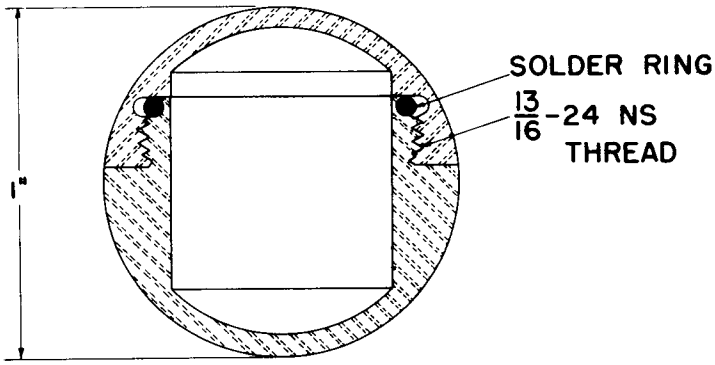


FIGURE 1

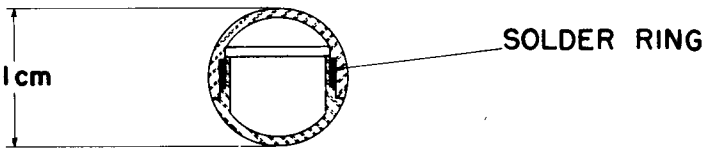
NEUTRON YIELD OF PLUTONIUM-BERYLLIUM ALLOYS



2a CYLINDRICAL SOURCE CAPSULE



2b SPHERICAL SOURCE CAPSULE



2c SPHERICAL SOURCE CAPSULE

FIGURE 2

NICKEL SOURCE CONTAINERS FOR $PuBe_{13}$

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