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A RADIATION BARRIER ALLOY FOR LONG-TERM STORAGE OF SPECIAL NUCLEAR MATERIALS: DEFINITION AND PRELIMINARY ASSESSMENT

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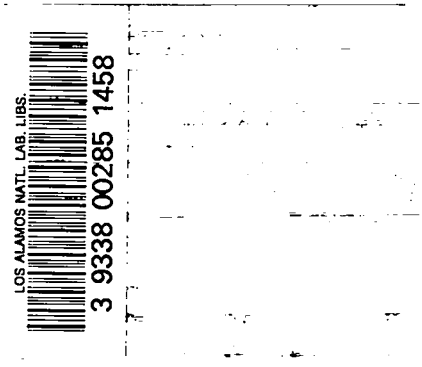

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Author(s): C. A. Beard, J. J. Buksa, J. W. Davidson, S. K. Lee, J. J. Park, J. W. Toevs, & K. A. Werley

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EXECUTIVE SUMMARY

The Radiation Barrier Alloy (RBA) concept is a method for introducing radioactive, chemical, and physical barriers for use in storage of weapons-grade plutonium, and yet still allow for accurate material control and accountability, as well as for retrieval of the material by the host nation if desired. The radioactive and chemical barriers are achieved by fabricating the plutonium in the form of a plutonium-beryllium compound ($PuBe_{13}$) which results in neutron emission resulting from (α,n) reactions within the compound and multiplication from (n,fission) processes in the plutonium. Pu-Be (α,n) neutron sources were developed as a replacement for Po-Be (α,n) neutron sources in the late 1940's and early 1950's. They have been in wide use ever since. The advantage of a Pu-Be source is that it is an intermetallic compound (as opposed to a mixture), and hence does not exhibit spectral effects due to grain size and heating effects. The conventional form is $PuBe_{13}$ with a density of 4.35 g/cm^3. Typical source sizes have been 0.01-0.02 m in diameter and 0.05-0.10 m in length resulting in emission of 10^6 - 10^8 neutrons/second. Pellets of this size can be placed in rods and arranged into arrays (similar to typical light-water-reactor fuel rod arrangements) to produce intense dose rates. Preliminary physics analyses have been completed, as well as a general review of fabrication techniques and availability of the required materials. These studies have resulted in the following conclusions:

- Dose levels in excess of 500 rem/hr at a 1-meter distance from the surface of the RBA assembly can be obtained.

- Essential for achieving these dose levels is operation at a high level of neutron multiplication ($k_{eff} \sim 0.9$).

- Criticality concerns, even under flooded conditions, can be eliminated through the use of a thermal-neutron-absorbing material (*e.g.*, cadmium) either as a cladding material or a container material surrounding the RBA assembly.

- Fabrication techniques for the Pu-Be compound are well demonstrated and fully compatible with the RBA assembly fabrication.

- Data from disassembly of Pu-Be sources indicate that the compound is stable and no significant physical degradation occurs over a 40-year time frame. There is no reason to believe that any additional problems exist for longer time frames, given that the components are designed for the appropriate lifetimes (*i.e.*, adequately account for gas production).

- The materials required for RBA implementation are available in the required quantities. Cost of these materials is not prohibitive. The possible exception is tantalum (see Section 4.1), although its use is non-essential for RBA performance and hence it will probably be eliminated from future RBA designs.

- Additional physical barriers can be added by welding the assembly together, and encasing the assembly in an outer container. If desired, the assembly (inside the outer container) can also be immersed in a neutronically-inert matrix such as lead. The lead serves a dual role in that in makes it difficult to move because of the additional weight, and it increases safety by reducing the possibility of a criticality accident resulting from flooding or assembly crushing.

To further the RBA preconceptual analyses, a baseline design based on physics performance was developed. A summary of the characteristics for this design is given in Table E-1. Based on this design, detailed thermal-hydraulic and structural stress calculations are being performed. In addition, an evaluation of the effects on the storage facility and a cost-benefit

analysis for the RBA approach are being conducted. For the baseline RBA configuration, approximately six RBA assemblies, each 31 m^3 in volume, would be required to store 50 MT of weapons-grade plutonium. A schematic representation of this is shown in Fig. E-1. This configuration results in a much smaller storage volume requirement than other storage options (such as storing as pits). Completion of these analyses will allow a better evaluation of the benefits of the use of RBA in the plutonium disposition process.

Table E-1. Characteristics of RBA baseline storage concept.

Array Size (Number of Pins)	36 X 36
Total Number of Pins	1296
Pu-Be Diameter (m)	0.032
Inner Clad (Tantalum) Thickness (m)	0.0025
Outer Clad (Stainless Steel) Thickness (m)	0.0025
Pitch (m)	0.09
Pitch-to-Diameter Ratio	2.14
Outer Can (Cadmium) Thickness (m)	0.01
Assembly Dimensions (m)	3.21 X 3.21 X 3.02
Assembly Volume (m^3)	31
Total Assembly Mass (kg)	45,900
Total Pu-Be Mass (kg)	13,600
Total Plutonium Mass (kg)	9,100
Total Tantalum Mass (kg)	17,500
Total Stainless Steel Mass (kg)	9,700
Total Cadmium Mass (kg)	5,200
Total (α,n) Neutron Source (n/s)	9.11e+11
k_{eff}	0.91
k_{eff} (flooded)	0.95
Dose 1 Meter from Side (rem/hr)	536
Dose 1 Meter from Top (rem/hr)	640

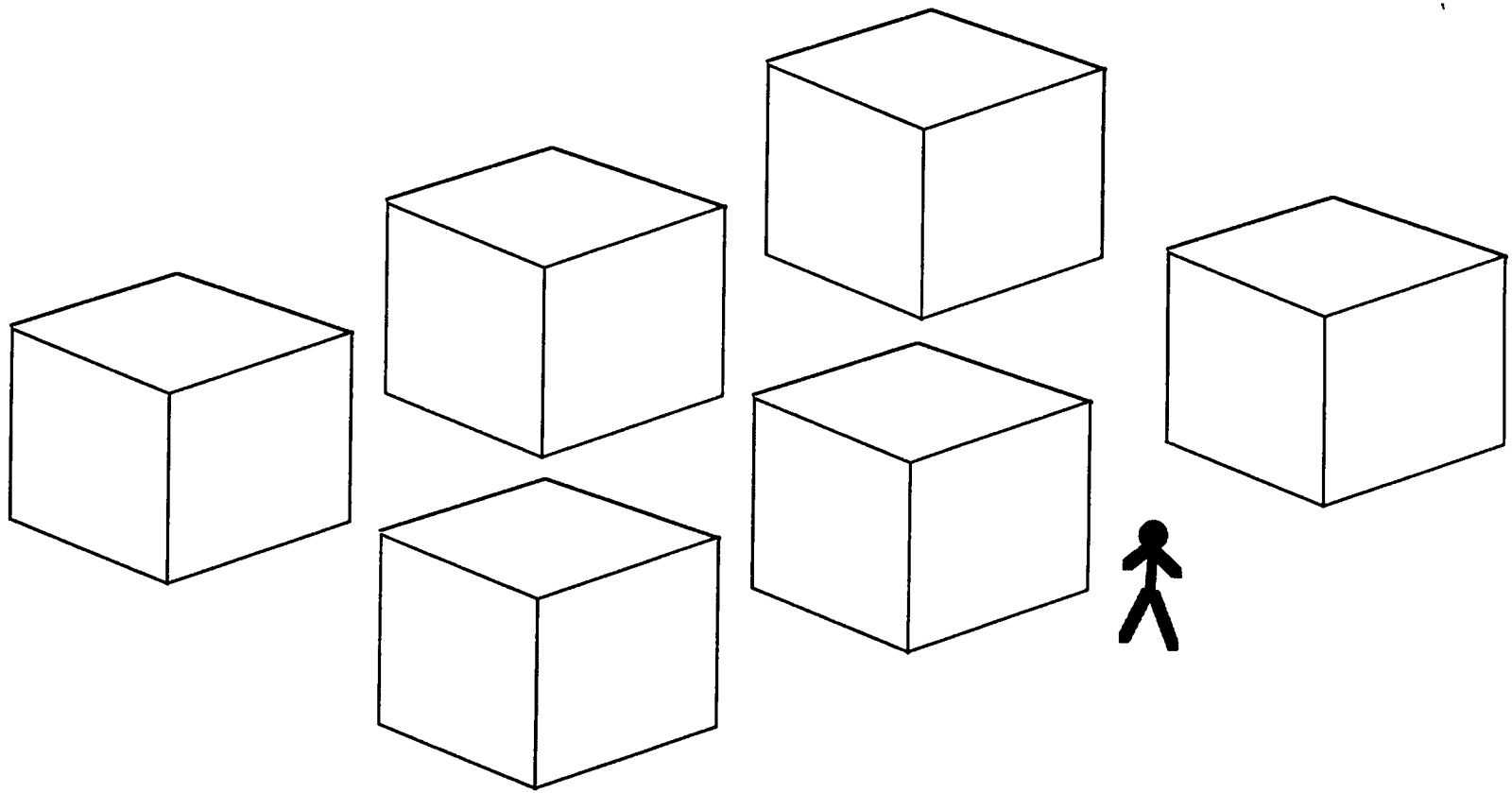

Figure E-1. Schematic representation of RBA storage.

1.0 INTRODUCTION

Dismantlement of warheads under the START treaties has resulted in increasing inventories of weapons-grade plutonium metal in both the United States and Russia which must be safely managed. Recently, the National Academy of Sciences (NAS) completed a study on the options for disposition of the weapons plutonium, and reached the following conclusions[1]:

- The United States and Russia should reach a reciprocal agreement that includes declarations of stockpiles of nuclear weapons and all fissile materials, cooperative measures to clarify and conform to those declarations, agree to halt the production of fissile materials for weapons, and agree to a monitored net reduction of the stockpiles.

- The United States and Russia should pursue a reciprocal regime of secure, internationally-monitored storage of fissile material, with the aim of ensuring that the inventory in storage can be withdrawn only for non-weapons purposes.

- The United States and Russia should pursue long-term disposition options that:

    a) minimize the time during which the plutonium is stored in forms readily usable for nuclear weapons;

    b) preserve material safeguards and security during the disposition process, seeking to maintain the same high standards of security and accounting applied to stored nuclear weapons (referred to as the "stored weapons standard");

    c) result in a form from which the plutonium would be as difficult to recover for weapons use as the larger and growing quantity of plutonium in commercial spent fuel (referred to as the "spent-fuel standard");

    d) meet high standards of protection for public and worker health and for the environment.

The NAS study identified the two most promising options for achieving these aims as: fabrication and use as fuel, without reprocessing, in existing or modified nuclear reactors; or vitrification in combination with high-level radioactive waste. However, the study recognized that implementation of the first option would require several decades at the minimum, including an initial ten years or so for the development of the fuel fabrication capabilities and licensing of current nuclear power plants to operate with the new fuel. With regard to the second option, Russia has made it clear that it considers plutonium an economic asset due to its energy production capability. Therefore, it is unlikely that Russia will consider any direct disposal methods such as vitrification. Finally, while the NAS study states that meeting the "spent-fuel standard" should be the first goal in long term disposition of the weapons plutonium, the panel also urges that means be pursued for going beyond the "spent-fuel standard", not just for the weapons plutonium, but for plutonium in spent fuel as well. A major factor behind this recommendation is the fact that the "spent-fuel standard" changes over time (with the decay of the spent fuel), and while spent fuel that has undergone decay for less than 100 years possesses a level of radioactivity that gives it a fair degree of proliferation resistance, the inherent resistance to diversion diminishes after that time.

From the NAS study, it is obvious that a method for increasing the resistance to diversion of the plutonium during storage before its final disposition is desirable. For this purpose the Radiation Barrier Alloy (RBA) concept was proposed, in which both radioactive and physical barriers would be employed, yet still allow accurate material control and accountability, as well as retrieval of the material if desired. In addition, the RBA concept provides a possible

mechanism to allow indefinite storage of materials, since for long-term disposition (>100 years), it meets and then ultimately exceeds the radioactivity component of the "spent-fuel standard". Finally, the RBA concept employs already developed technologies which would allow full implementation within ten years.

1.1 THE SPENT-FUEL STANDARD

As stated above, the NAS study concluded that the disposition concept selected should result in a form from which the plutonium would be as difficult to recover for weapons use as the larger and growing quantity of plutonium in commercial spent fuel. This was referred to as the "spent-fuel standard". The reason behind this conclusion was twofold: 1) if the selected option left the material in a form less resistant to proliferation than spent fuel, a special class of nuclear material would still exist and thus require special safeguards; and 2) selecting an option which made the material more resistant to proliferation did not make sense unless the much greater inventory of plutonium contained in spent fuel was also incorporated into the concept. Having established the "spent-fuel standard" as the reference point for disposition options, the NAS study did not give a concrete definition of what was required to meet this standard. However, the study did give four primary factors which affect the usefulness of civilian spent fuel as a potential weapons material: a) the intense radioactivity of the fission products in the fuel (which decays with time); b) the need for chemical separation of the plutonium from the fuel (which must be accomplished by remotely-operated equipment as long as the fuel remains intensely radioactive); c) the isotopic composition of the plutonium; and d) if the party in question does not own the spent fuel, the difficulty of acquiring it. With regard to proliferation-resistance, the main advantage of (a) is the requirement for remote handling facilities to manipulate the material. The advantage of (b) is that chemical processes are difficult and cumbersome if they must be performed remotely. While (c) is a minor advantage because the isotopic composition of spent-fuel is not as ideal as that produced specifically for weapons, it is not a major deterrent because a nuclear device can still be constructed using the isotopic mix found in normal light-water reactor spent-fuel discharge unless a special fuel cycle is used which produces large amounts of ^{238}Pu whose heat generation rates can cause significant fabrication difficulties. Finally, (d) is dependent solely on the material location, physical safeguards, and the associated size and weight of the assemblies. Characteristic dimensions and weights of typical reactor fuel assemblies are given in Table 1-1.

Although RBA is not currently envisioned as a final disposition form (although this could still be a possibility), satisfying the "spent-fuel standard" is still desirable to provide a high-level of diversion resistance for the stored material and a deterrent to possible theft. Therefore, RBA performance requirements were set to approximate this standard. In lieu of a well-defined "spent-fuel standard", the proliferation resistant characteristics outlined above (with the exception of the isotopic distribution which is not a large contributor to the proliferation resistance of the material) can be satisfied by meeting the following criteria:

1) remote handling is required;

2) chemical separation is required for plutonium recovery; and

3) the size and weight of the final form is comparable to or larger than a spent-fuel assembly.

If these three conditions are met, the material is effectively as proliferation resistant as spent-fuel and hence approximates the "spent-fuel standard".

To meet the remote handling criteria, the dose rate emitted must be sufficient to induce radiation sickness. As shown in Fig. 1-1, radiation-exposure-syndrome begins at a whole-body dose of 100 rem, therefore remote handling is required at this dose or above. However, the dose

Table 1-1. Characteristics of light-water reactor fuel assemblies.

Reactor	Array Size	Dimensions (mm)	Length (m)	Weight (kg)
PWR* (a,b)	17 x 17	217.0 x 217.0	4	652
System 80+ (c,d)	16 x 16	202.7 x 202.7	4.13	651.36
BWR** (a)	8 x 8	139.0 x 139.0	4.1	273
ABWR (e)	8 x 8	139.0 x 139.0	4.1	273
CANDU (a,f)	37 elements	102 diameter	4.95	22.5

* Babcock & Wilcox

a-Nuclear Systems I, Todreas & Kazimi

b-Nuclear Engineering, 2nd Ed., Knief, 1993, p. 708

c-DOE Plutonium Disposition Study, Pu Consumption in ALWRs, ABBCE, May 15, 1993.

d-System 80+ Standard Design, Cessar Design Certification, Volume 4.

** General Electric BWR/6

e-Study of Plutonium Disposition using the GE Advanced Boiling Water Reactor

f-AECL/DOE Plutonium Dispositioning Study Mid-Term Review, June 7, 1994.

administered is a function of the time, distance, and shielding incorporated in the diversion effort. Both the NRC and IAEA consider material emitting 100 rem/hr at 1 meter to be sufficiently resistant to diversion that the required safeguards category can be reduced. If material was to be transported in the form of RBA, a minimum of 100 rem/hr would provide for some self-protection and allow the safeguards category to be reduced. However, since storage of the material is for a greater length of time, providing more opportunity for a well-planned and organized diversion effort, dose rates which produce a greater level of passive protection (similar to that emitted by spent-fuel) are desirable. Figure 1-2 shows the time to the onset of the prodromic phase of radiation sickness (nausea, fatigue, vomiting, *etc.*) as a function of dose rate and exposure time. For ten-year-old spent-fuel (dose rate equal to ~2000 rem/hr at 1 meter), the exposure time required to cause radiation sickness is on the order of minutes. To achieve a comparable required exposure time for RBA, a minimum 1-meter dose rate on the order of 400-500 rem/hr is required.

The chemical separation criteria is met by the fact that the plutonium in the RBA assemblies is in the form of $PuBe_{13}$ compound, from which the plutonium cannot be removed except through chemical means. This involves dissolution of the compound in a hydrochloric acid solution and then subsequent processes to convert the plutonium into a pure metallic form.

The size and weight criteria are easily met, even for an RBA assembly designed to be transported. Additional physical barriers can also be incorporated into the RBA storage assembly design including encompassing the entire assembly in an outer container and/or immersing the entire assembly in a neutronically-inert material such as lead.

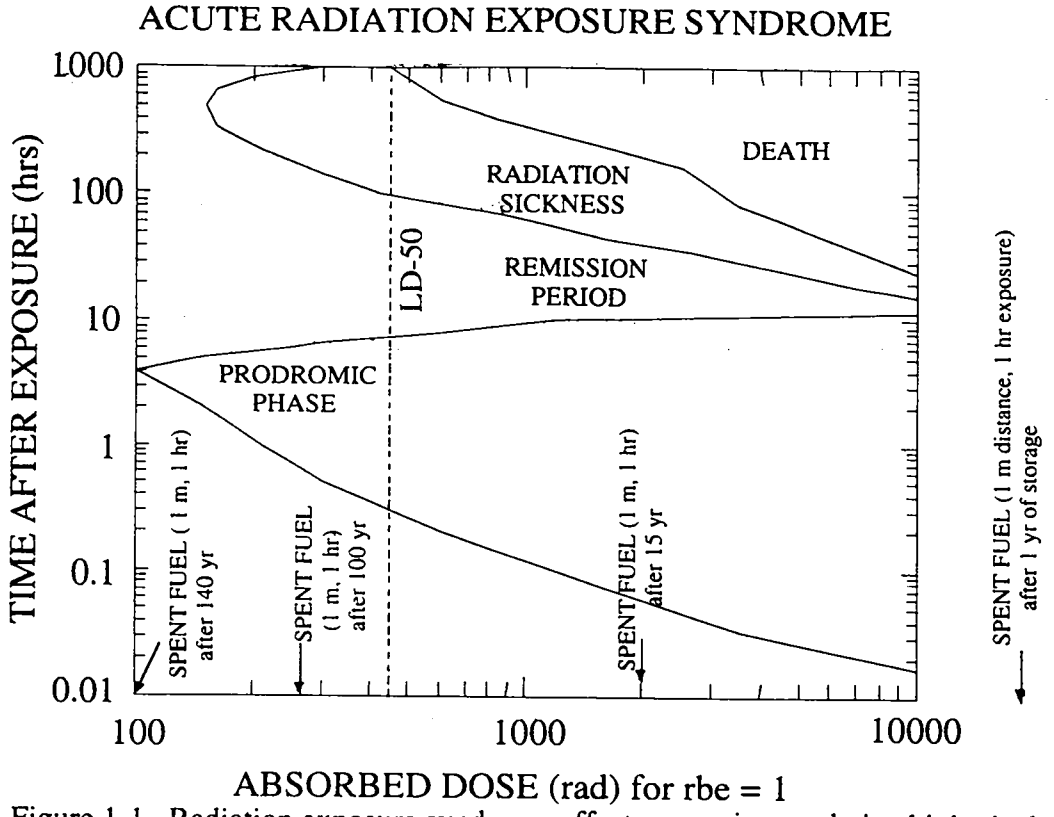

Figure 1-1. Radiation exposure syndrome effects assuming a relative biological effectiveness(rbe) equal to 1.

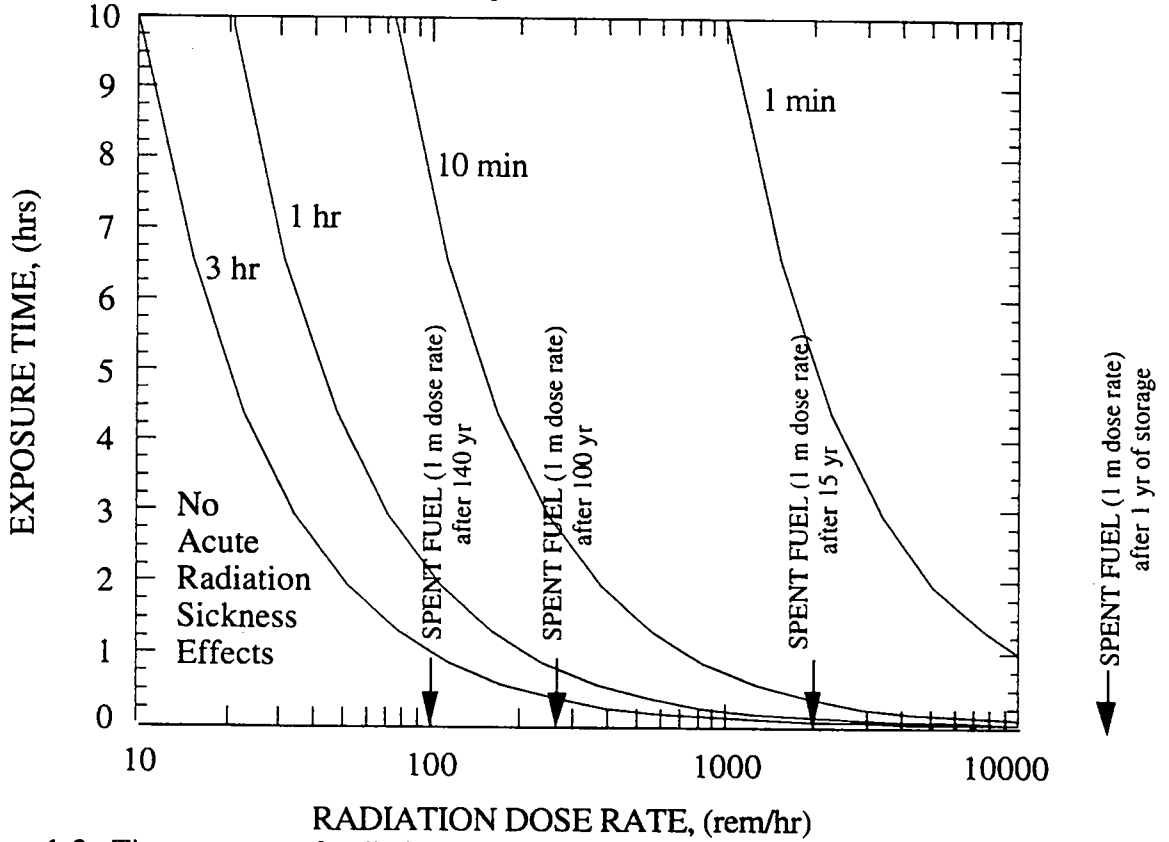

Figure 1-2. Time to onset of radiation exposure syndrome prodromic phase (nausea, vomiting, fatigue, *etc.*) as a function of dose rate and exposure time.

2.0 OVERVIEW OF THE RBA CONCEPT

The RBA concept provides radioactive, chemical, and physical barriers to the diversion of weapons-grade plutonium. The radioactive and chemical barriers are achieved by fabricating the plutonium in the form of a plutonium-beryllium alloy matrix ($PuBe_{13}$) which results in intense dose rates due to neutron emission resulting from (α,n) reactions within the matrix and multiplication from (n,fission) processes in the plutonium.

Pu-Be (α,n) neutron sources were developed as a replacement for Po-Be (α,n) neutron sources in the late 1940's and early 1950's. They have been in wide use ever since. The advantage of a PuBe source is that it is an intermetallic compound (as opposed to a mixture), and hence does not exhibit spectral effects due to grain size and heating effects. The conventional form is $PuBe_{13}$ with a density of 4.35 g/cm^3. Typical source sizes have been 0.01-0.02 m in diameter and 0.05-0.10 m in length resulting in emission of 10^6 - 10^8 neutrons/second. Pellets of this size can be placed in rods (similar to typical fuel rod arrangements) to produce more intense dose rates.

If it is desired to recover the plutonium or terminate the activity, chemical separation of the Pu from the Be leaves only the original radiological hazard of the Pu itself and a small amount of fission products produced through fission reactions in the RBA matrix. The RBA form may itself be acceptable for repository disposition, although this is uncertain at this time. If not, conversion to a suitable form for geologic disposal may be accomplished, as above, with no radiological concerns other than that of the original plutonium and the minimal amount of fission products generated. It should also be possible to leave the beryllium in metallic form as metal buttons, avoiding the chemical hazard of beryllium as a fine particulate.

2.1 MISSION DEFINITION: RBA APPLICATIONS

The currently envisioned plan for the disposition of stockpile plutonium is shown in Fig. 2-1. The plan is divided into two phases: the initial phase would involve disassembly of the pits, recovery of Pu residues from various facilities, and the placement of this material into interim storage. The second phase would involve the removal of this material from storage and either its use for nuclear power generation in conventional light-water reactors (LWRs) as MOX fuel and placement of the residual wastes (including any plutonium remaining in the spent-fuel) in a repository, or disposal in a geologic medium which could involve vitrification with high-level waste and placement in a repository, placement in a deep bore hole, or just vitrification and placement in a repository. A third alternative exists, called the "no-action" alternative, in which the plutonium would continue to be stored (although at a different site than the initial interim storage) indefinitely until a decision was made as to the mechanism of its ultimate disposal. This option allows longer-term concepts such as Accelerator-Based Conversion (ABC) or deep-burn reactors to be considered, although ultimately geologic disposal or single-cycle burn may still be selected.

The RBA concept is a mechanism to provide a passive barrier to diversion if the no-action alternative is selected. This application allows extension of the timelines required to make a decision with regard to ultimate disposition because the material is self-protecting with a very long half-life (24000 years).

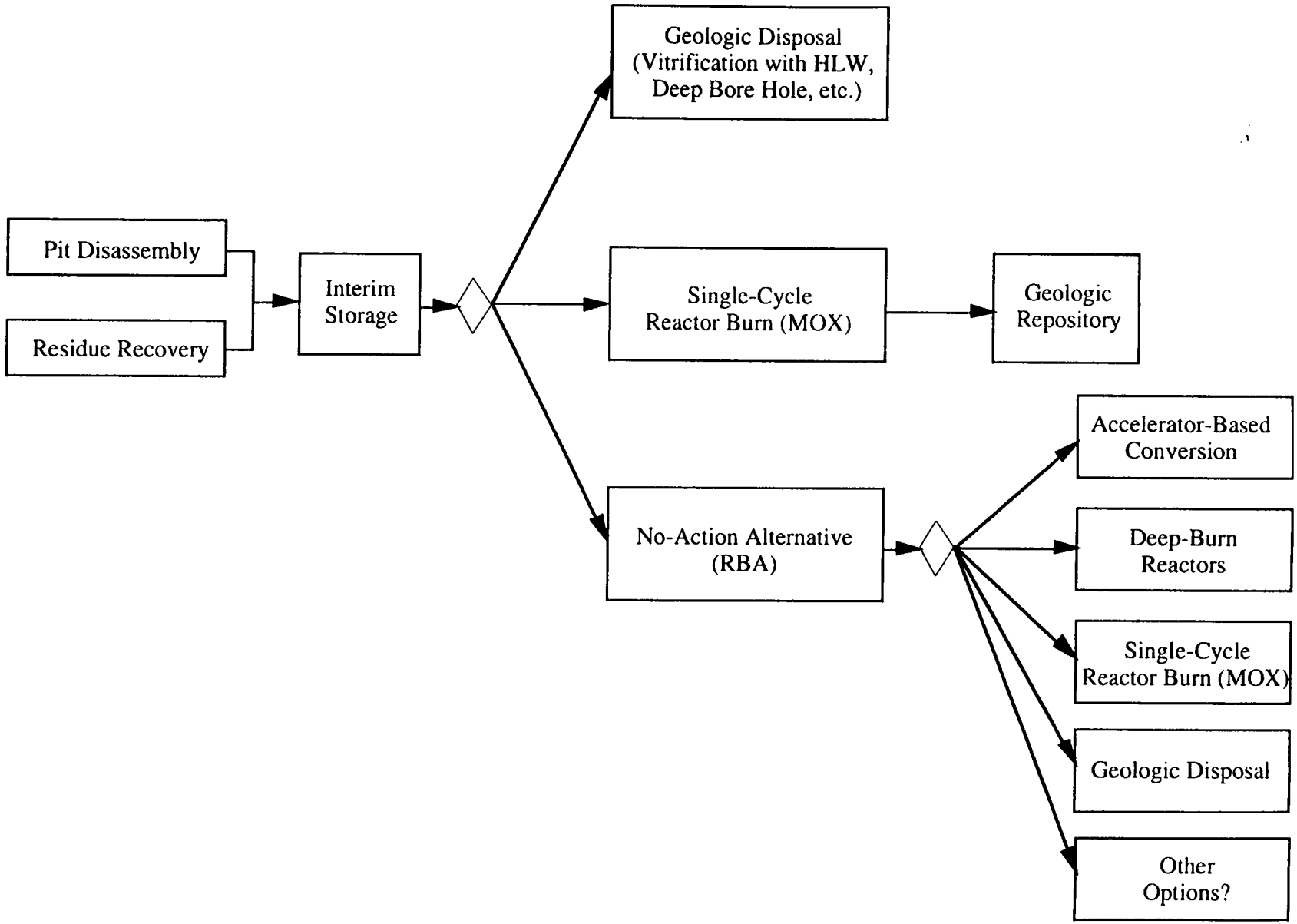

Figure 2-1. Plutonium disposition plan with RBA option.

3.0 RBA REQUIREMENTS

Specific criteria have been developed from general (high-level) requirements. These general requirements for the RBA concept are as follows:

- Safety - RBA must meet all applicable safety criteria during fabrication, transportation, storage, and ultimate removal. Risks to the public and work force must be minimized.

- Fabrication - Fabrication of the alloy and associated structural material must be feasible in the required size and configuration needed to meet the concept criteria. Also, the fabrication process must occur at a sufficient rate to place the desired amount of material in storage within a reasonable time frame.

- Environmental protection - The concept must not have large negative environmental impacts.

- Resistant to removal - The material must be resistant to unauthorized removal from the storage material.

- Resistant to diversion - The material must be resistant to diversion during fabrication and transportation prior to placement of the material in the storage facility.

- Cost-effective - The cost of the fabrication and storage process must not be prohibitively high.

- Materials control & accountability (MC&A) - The concept should possess the ability to track the plutonium inventory in storage.

- Feasible for storage over the desired time period - The concept should allow storage for the time period desired in a safe and reliable manner.

- Russian acceptability/buy-in - The concept should be synergistic with Russian nuclear materials management goals and have a significant chance for concurrent implementation between Russia and the United States.

- Near-term - The concept should be able to be applied in the near-term in comparison to other storage options.

- Recoverable by host nation - The plutonium should be recoverable in a safe and efficient manner by the host nation.

To achieve each of these general requirements, a set of specific criteria must be met.

- Safety - Exposure to nuclear workers from normal conditions shall be as low as reasonably achievable and in no case exceed 5 rem/yr. Exposure to the general population from normal conditions shall be as low as reasonably achievable and in no case exceed 0.5 rem/yr. Exposure of workers to toxic materials shall be as low as practical and in no case exceed EPA guidelines. Releases of toxic materials to the environment shall be as low as reasonably achievable and in no case exceed EPA general guidelines at the site boundary.

- Fabrication - Fabrication of RBA assemblies must be possible at a rate equal to or greater than the rate of pit disassembly. Pit disassembly at a rate of approximately 1000 pits/yr is anticipated.

- Environmental protection - The RBA concept must have a satisfactory Programmatic Environmental Impact Statement (PEIS). No severe environmental consequences can result from any stage in the RBA process including fabrication, transportation, storage, or removal.

- Resistant to removal/diversion - In lieu of any well-defined "spent-fuel standard", a single RBA assembly shall meet the following characteristics:

  - emission of a radiation barrier in excess of 400 rem/hr at 1 meter in any direction from the surface of the assembly during storage.

  - emission of a radiation barrier in excess of 100 rem/hr at 1 meter in any direction from the surface of the assembly during transportation (if transportation in the form of RBA is desired).

  - a minimum combined length plus girth of 454 cm which is equivalent to the smallest LWR fuel assembly (BWR, Table 1-1).

  - a minimum mass of 273 kg which is equivalent to the smallest LWR fuel assembly (BWR, Table 1-1).

  - the RBA will be in the form of the intermetallic alloy $PuBe_{13}$ (perhaps with $AmBe_{13}$, $NpBe_{13}$, *etc.)* which will be placed inside a sealed package material. Separation of the special nuclear material from the sealed package and internal matrix shall require multiple physical and chemical processes.

- Storage - The final storage form shall be safe and robust and allow for reliable storage over an extended period of time. Criticality will be prevented under all anticipated conditions. Heat removal from the assemblies shall be accomplished solely through passive mechanisms, and no active gas removal systems will be incorporated into the facility. Required remote facilities will be identified.

- Cost effective - The RBA concept must cost less than alternative options for either interim/long-term storage and/or disposition, or possess identifiable benefits which justify any additional cost.

- Materials control & accountability - The RBA process shall allow accountability at all stages. Unobserved removal of material shall be made prohibitively difficult. The RBA will be compatible with traditional safeguards and security methods used to monitor and detect the movement of materials or sealed packages.

- Feasible for long-term storage - RBA assemblies designed for long-term storage shall be designed to maintain their physical integrity as well as the removal/diversion resistance criteria outlined above for a period of time to be defined by national policy objectives.

- Russian acceptability/buy-in - The RBA concept shall be synergistic with Russian goals in that the special nuclear material shall be available for recovery for future use if desired. The process will not have prohibitive costs which preclude Russian implementation. In addition, the RBA concept will not involve any technologies that would be unsuitable for transfer to Russia.

- Near-term - The RBA concept shall be fully designed and available for implementation within 3 years. Full implementation will be possible within 10 years.

- Recoverable by host nation - The special nuclear material shall be recoverable, but only in a controlled and easily observable manner. Recovery of the material shall not prevent the continuation of MC&A practices for both the material being removed as well as that remaining in storage.

4.0 RBA FABRICATION

Two proven fabrication techniques have been identified for the manufacturing of Pu-Be sources. The first involves mixing the plutonium and beryllium as powders in the proper stoichiometric ratios (33% beryllium, 67% plutonium by weight), and heating the mixture to 1100 °C (with exothermic spikes to approximately 1400 °C) which creates the compound $PuBe_{13}$. To convert the material into the proper phase for optimal neutron production, the material must then be reheated to 2000 °C where it is entirely in the liquid phase and then cooled to form a solid. The second fabrication technique involves melting the plutonium and then dissolving beryllium in the proper amount into the plutonium liquid. In this case, it is unnecessary to convert the beryllium into powder, and thus the airborne hazard of beryllium powder can be avoided. The largest sources made using these techniques contain between 100-200 grams of plutonium. Therefore, to form an RBA pin to place in an assembly, the Pu-Be must be fabricated in the form of pellets and then placed into a clad. Because high temperatures are required during the Pu-Be fabrication, a high-temperature crucible is required that is compatible with plutonium Traditionally, tantalum has been used for this purpose, and the current assumption is that it will also be used in RBA, although alternate materials are being investigated Whatever material is used for the crucible is assumed at this point to serve also as the first clad material (it will be sealed upon completion of the Pu-Be fabrication) which would then be placed inside the rod cladding, thus providing for double-containment of the Pu-Be. In conventional Pu-Be source, stainless steel is used for the outer cladding material, and thus is currently being used for the baseline RBA configuration. The rods will then be arranged in the appropriate manner using spacers and welded into place inside an outer container which is currently envisioned as cadmium. If immersion in a lead matrix is desired, molten lead will be poured over the completed assembly inside the outer container and allowed to solidify. The outer container will then be sealed.

Automation of the fabrication process and the remote facilities required to configure the sources into arrays of rods is currently being investigated. Fabrication of a cadmium can to surround the Pu-Be rod array is possible, but the cadmium thickness is limited to approximately 1/8 inch in thickness. Also, cadmium cannot be welded in air because it is flammable, so another method will have to be used for sealing the can, or an additional container which can be welded will have to surround the cadmium. Details with regard to assembly of the full array will be completed upon further analysis and engineering of the baseline RBA design.

4.1 MATERIAL AVAILABILITY

The availability of the beryllium required for RBA has been investigated. Approximately half of the amount required will be available from the disassembled pits, and there appears to be no problem in obtaining the remainder (large inventories have already been volunteered from other institutions if desired). The availability of other materials which might be required have also been studied, and the results indicate that while there are insufficient inventories of ^{238}Pu or ^{241}Am to use as neutron-source enhancers (see section 5.8.1), cadmium is obtainable for use as an outer can at a price of $13 per pound, tantalum is obtainable at a price of $120 per pound, lead is obtainable at a price of $0.55 per pound, and stainless steel is obtainable at a price of $1.35 per pound. Table 4-1 shows the amount of materials required for the baseline RBA design outlined in section 5.9 and the associated costs of obtaining these materials. The large amount of tantalum required could impact its market value based on availability quoted by current tantalum vendors (one supplier quoted a maximum supply rate for tantalum of 630 kg every 5 weeks). This issue,

coupled with its high cost, will probably eliminate tantalum from future RBA assembly designs. However, its presence is not essential, but was just assumed due to its historical use in Pu-Be sources. Even if a suitable replacement for tantalum cannot be found for use as a crucible in the Pu-Be fabrication process, the Pu-Be can be removed from the tantalum upon solidification and clad in any material desired, and the small amount of tantalum required for the fabrication process can be reused.

Table 4-1. Material costs associated with the baseline RBA design.
(Approximately 6 assemblies required for 50 MT of plutonium)

Material	Required Mass (kg)	Price per kg	Total Cost
Plutonium	50,000	$0.00	$0
Beryllium	24,725	$0.00[1]	$0
Cadmium	28,571	$28.60	$817,131
Tantalum[2]	96,153	$264.00	$25,384,392
Stainless Steel	53,297	$2.97	$158,292
Lead (If desired)	1,615,400	$1.21	$1,954,634
Total			$28,136,766

[1] Required beryllium is assumed to come from DOE complex inventories (disassembled pits plus additional available inventories)

[2] Tantalum will probably be removed from future RBA designs due to its high cost and potential availability problems. Its presence is not essential for RBA performance.

4.2 SYNERGISM WITH THE PU-BE SOURCE DISASSEMBLY PROGRAM

Currently, a program exists at Los Alamos National Laboratory for the recovery and disassembly of Pu-Be sources that were manufactured during the last forty years. Current data from disassembly of these Pu-Be sources indicate that the compound is stable and no significant physical degradation occurs over this time frame. There are no current indications that any additional problems exist for longer time frames, given that the components are designed for the appropriate lifetimes (*i.e.* adequately account for gas production). Additional information can be obtained by designing RBA-specific experiments involving the Pu-Be sources to be disassembled. These experiments can included more detailed materials testing and characterization, as well as using the available sources to construct Pu-Be arrays comparable to RBA configurations to verify criticality and dose calculations. The availability of these sources will allow the generation of a substantial experimental data base for RBA that otherwise would require a much greater time frame and funding level to compile.

5.0 PHYSICS ANALYSES

5.1 CALCULATIONAL METHODOLOGY

Pu-Be (α,n) neutron sources are typically fabricated in the form $PuBe_{13}$ which is an intermetallic compound with a typical density of 4.35 g/cm^3.[2,3] This is the form used in all the calculations described here. When americium is present, it is assumed to displace the plutonium, but still exist in the form of $AmBe_{13}$ and have a negligible effect on the material's mass density. The three-dimensional, Monte Carlo transport code MCNP[4] was used to perform all of the calculations and the 1977 ANSI/ANS[5] conversion factors were used for flux-to-dose rate conversion. These conversion factors (with the associated quality factors for the neutrons) are

shown in Tables 5-1 and 5-2. The neutron source was input as a homogeneous source uniformly distributed throughout the Pu-Be region. The energy spectrum for the neutron source was taken from reference (6) and Fig. 5-1 displays the group structure in which it was input into MCNP. Since MCNP begins with source neutrons, and does not deal with the (α,n) reaction itself, a method is required for determining the (α,n) neutron source strength (n/s) which is generated by any Pu-Be configuration. For the calculations, the neutron yield was taken to be 6.7e+07 n/s/kg Pu-Be.[3]

5.2 BENCHMARKS

To estimate the accuracy of the calculations, comparisons were performed with experimental results quoted in reference (6). Experimental measurements of neutron source strength and normalized dose (dose per unit fluence) were given for four Pu-Be sources of different sizes. The physical compositions (including the isotopic mixtures of the plutonium) of the sources are given in Table 5-3.

Using the Pu-Be density and neutron yield stated above and taking into account the isotopic distribution and the fact that the specific activity of ^{240}Pu is 3.7 times that of ^{239}Pu (^{241}Pu decays predominantly by beta and hence has an insignificant effect on the initial source strength; however, the production of ^{241}Am by beta decay from ^{241}Pu does result in an increase in (α,n) source strength over time), the source strength for each sample was calculated. Table 5-4 shows these numbers as well as the experimental values.

To perform the dose comparisons, MCNP models of the sources were created and the doses calculated at various distances from the samples. Each dose was then normalized per unit fluence which resulted (as expected) in a spatially independent (no statistically significant fluctuations) value for each source. These values as well as the experimental values are shown in Table 5-5. It should be noted that an additional reference[7] quoted a normalized Pu-Be dose value of 3.52e-08 rem/n/cm^2.

From Tables 5-4 and 5-5, it can be seen that the predictions with regard to the neutron source are 1.5% to 11.2% in excess of those determined experimentally, while the dose calculations are 3.0% to 6.1% lower than the experimental values. Therefore, the physics calculations modeling RBA assemblies can be assumed to be accurate within these bounds.

5.3 CALCULATIONAL PHILOSOPHY

The RBA calculations performed to date can be separated into two distinct categories. The first category was a series of parametric calculations to determine the relationship between dose rates and multiplication and to predict the maximum dose rates achievable by small, "fuel-assembly-like" RBA arrays (Sections 5.4-5.7). While the results of these calculations are useful in defining the relationships between the major parameters (size, geometry, multiplication, *etc.*) in determining RBA performance, the assemblies analyzed fail to achieve the dose rates required for a long-term storage application (although some of the assemblies are suitable in size and dose emission for a transportation application). Consequently, the second series of calculations (Sections 5.8-5.9) concentrate on increasing the dose rates to those required for storage, and then defining a baseline storage concept to allow further engineering analyses. The baseline storage concept generated is not suitable for transportation due to its size, and so if transportation in the form of RBA is desired, an appropriate coupling between small assemblies (emitting around 100 rem/hr) and the final storage form will be required.

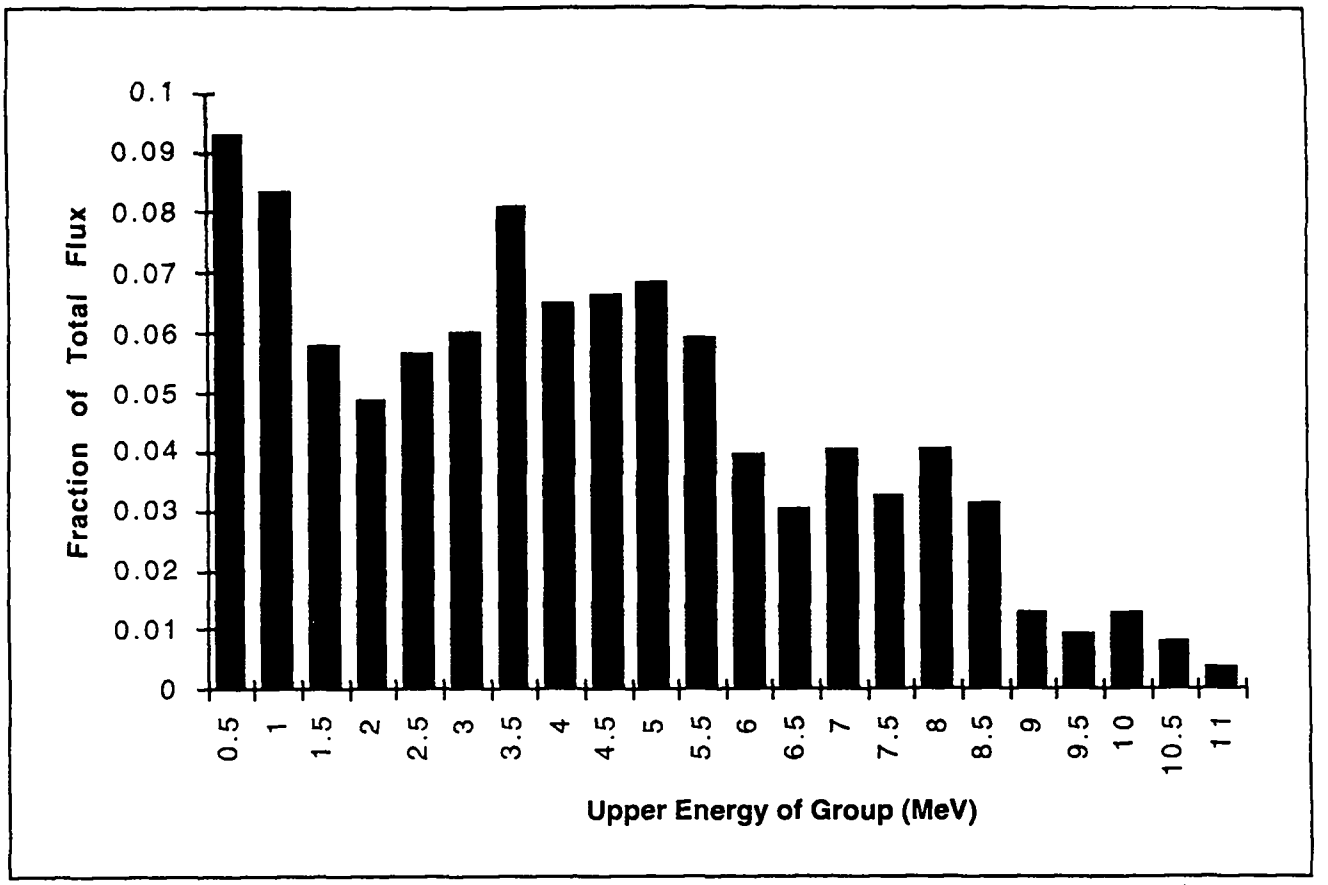

Figure 5-1. Energy spectrum of Pu-Be (α,n) neutrons used for transport calculations.[6]

Table 5-1. ANSI/ANS-1977[5] neutron flux-to-dose rate conversion factors.

Neutron Energy (MeV)	(rem/hr)/(n/(cm^2-sec))	Quality Factor
2.5e-08	3.67e-06	2
1.0e-07	3.67e-06	2
1.0e-06	4.46e-06	2
1.0e-05	4.54e-06	2
1.0e-04	4.18e-06	2
1.0e-03	3.76e-06	2
1.0e-02	3.56e-06	2.5
1.0e-01	2.17e-05	7.5
5.0e-01	9.26e-05	11
1.0	1.32e-04	11
2.5	1.25e-04	9
5.0	1.56e-04	8
7.0	1.47e-04	7
10.0	1.47e-04	6.5
14.0	2.08e-04	7.5
20.0	2.27e-04	8

Table 5-2. ANSI/ANS-1977[5] gamma flux-to-dose rate conversion factors.

Gamma Energy (MeV)	(rem/hr)/(gamma/(cm^2-sec))
0.01	3.96e-06
0.03	5.82e-07
0.05	2.90e-07
0.07	2.58e-07
0.1	2.83e-07
0.15	3.79e-07
0.2	5.01e-07
0.25	6.31e-07
0.3	7.59e-07
0.35	8.78e-07
0.4	9.85e-07
0.45	1.08e-06
0.5	1.17e-06
0.55	1.27e-06
0.6	1.36e-06
0.65	1.44e-06
0.7	1.52e-06
0.8	1.68e-06
1.0	1.98e-06
1.4	2.51e-06
1.8	2.99e-06
2.2	3.42e-06
2.6	3.82e-06
2.8	4.01e-06
3.25	4.41e-06
3.75	4.83e-06
4.25	5.23e-06
4.75	5.60e-06
5.0	5.80e-06
5.25	6.01e-06
5.75	6.37e-06
6.25	6.74e-06
6.75	7.11e-06
7.5	7.66e-06
9.0	8.77e-06
11.0	1.03e-05
13.0	1.18e-05
15.0	1.33e-05

Table 5-3. Experimental Pu-Be source characteristics[6] used for calculation benchmarks.

Source name	Mass Pu (g)	Mass Be (g)	Atom fraction Pu^{239}	Atom fraction Pu^{240}	Atom fraction Pu^{241}
M-99	2.0	1.1	0.97	0.03	0.001
M-977	13.5	6.6	0.92	0.07	0.007
M-591	79.8	39.0	0.95	0.05	0.004
M-930	159.9	79.0	0.92	0.07	0.007

Table 5-4. Comparison of calculated and experimental[6] Pu-Be source strengths.

Source name	Experimental Source Strength (neutrons/sec)	Calculated Source Strength (neutrons/sec)	Calc./Exp.
M-99	2.0e+05	2.24e+05	1.120
M-977	1.6e+06	1.64e+06	1.025
M-591	8.9e+06	9.03e+06	1.015
M-930	1.8e+07	1.95e+07	1.083

Table 5-5. Comparison of calculated and experimental[6] normalized dose values.

Source name	Experimental Dose ($rem/n/cm^2$)	Calculated Dose ($rem/n/cm^2$)	Calc./Exp.
M-99	3.76e-08	3.56e-08	0.947
M-977	3.63e-08	3.52e-08	0.970
M-591	3.55e-08	3.43e-08	0.966
M-930	3.60e-08	3.38e-08	0.939

5.4 ARRAY-SIZE PARAMETRIC CALCULATIONS

Initial parametric calculations consisted of arrays of 1-m long, 0.028-m diameter $PuBe_{13}$ pins double-clad with 0.0025-m thick tantalum and 0.0025-m thick stainless steel. The pins are arranged in a square lattice with a pitch-to-diameter (including clad) ratio of 1.1. Table 5-6 lists assembly characteristics including the array sizes tested, the corresponding total assembly mass and Pu mass, the total (α,n) neutron source emitted, and the effective multiplication factor for the bare assembly. Dose rates were obtained at the axial mid-plane of the assembly at 0.5-m, 0.75-m, 1-m, 1.25-m, and 1.5-m distances from the center of the assembly. These values are shown in Fig. 5-2. In addition dose rates were calculated at a 1-m distance from the surface of the assembly at the axial mid-plane as well as a 1-m distance from the top of the assembly at the centerline. Figure 5-3 displays these results. As expected, the calculations indicate a decrease in dose as the location moves away from the assembly and an increase in dose with increasing array size. Figure 5-4 plots the dose rates at 1 meter from the surface of the assembly as a function of assembly multiplication ($1/(1-k_{eff})$). It can be seen from Fig. 5-4 that the relationship between dose and multiplication is nearly linear, indicating that multiplication effects are dominant over any addition to dose due to increasing (α,n) neutron source strength. This is further illustrated in Fig. 5-5 which shows the same dose rate as a function of plutonium mass; this relationship is clearly non-linear which would not be the case if the observed increase in dose for the larger array sizes was due mostly to the increase in the (α,n) neutron source strength.

Table 5-6. Array characteristics for tantalum/stainless-steel-clad pins.

Number of Pins in Array	13X13	14X14	15X15	16X16	17X17	18X18	19X19	20X20
Assembly Width (cm)	53.96	58.14	62.32	66.50	70.68	74.86	79.04	83.22
Total Assembly Mass (kg)	1503.07	1743.20	2001.12	2276.84	2570.34	2881.63	3210.71	3557.57
Mass Pu (kg)	303.79	352.32	404.45	460.17	519.49	582.40	648.91	719.02
Total Neutron Source (n/sec)	3.03E+10	3.52E+10	4.04E+10	4.59E+10	5.19E+10	5.81E+10	6.48E+10	7.18E+10
Keff	0.736	0.777	0.808	0.839	0.861	0.889	0.905	0.924

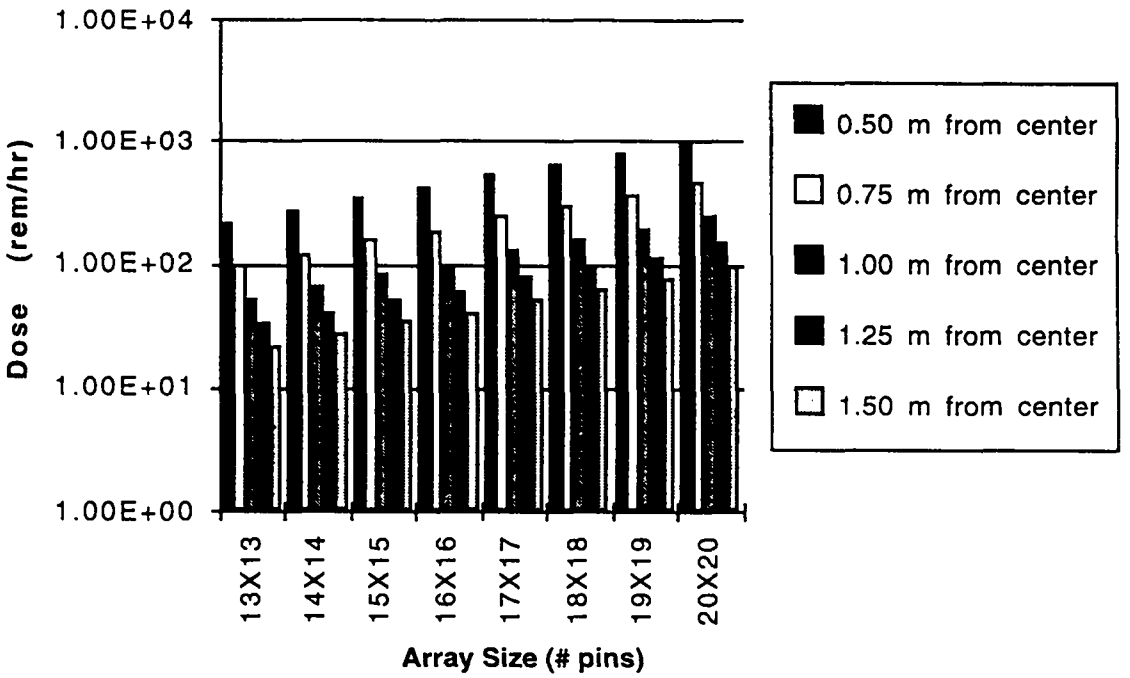

Figure 5-2. Dose rates at axial mid-plane of assembly of tantalum/stainless-steel-clad pins.

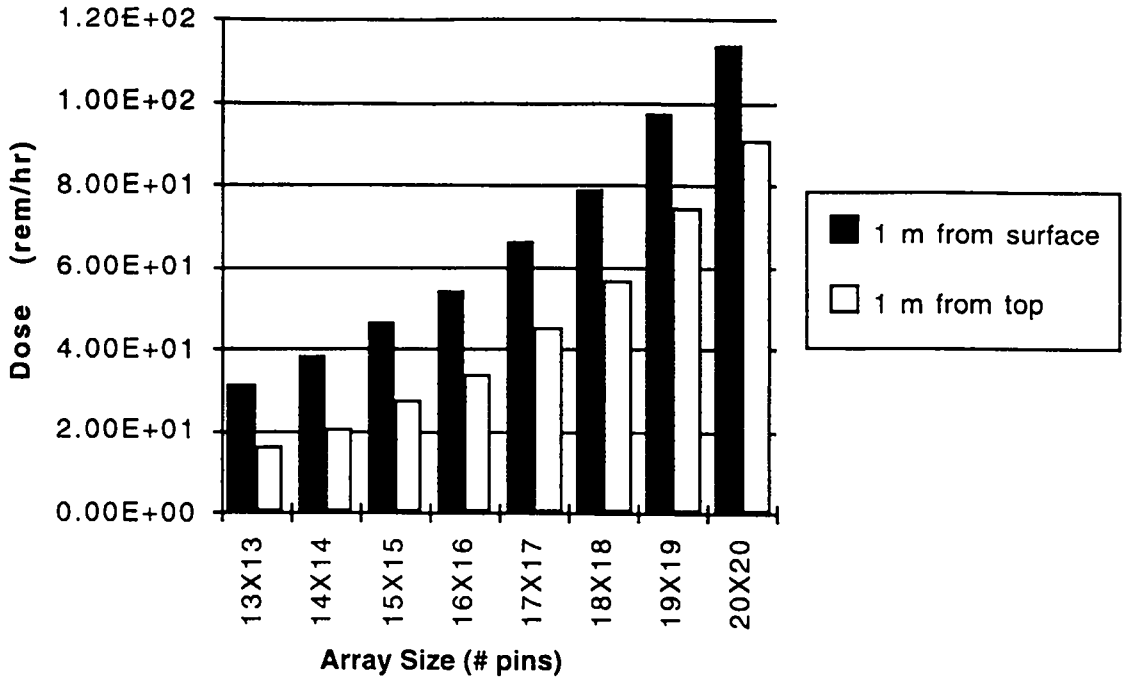

Figure 5-3. Dose rates 1 meter from surface (at axial mid-plane) and 1 meter from top (on centerline) for tantalum/stainless-steel-clad pins.

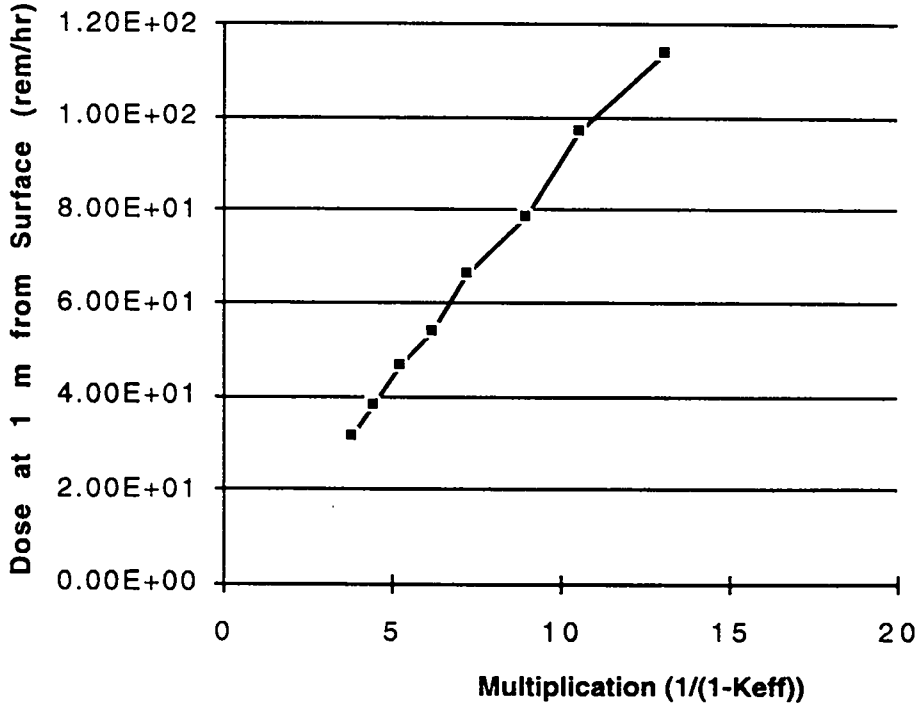

Figure 5-4. Dose rate 1 meter from surface (at axial mid-plane) as a function of multiplication.

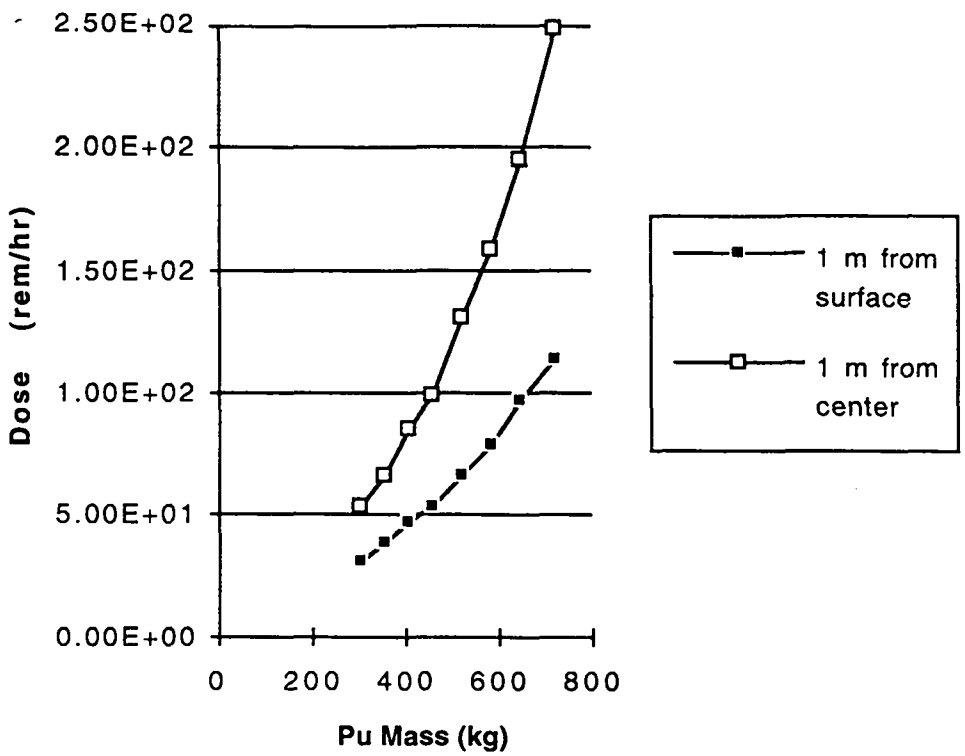

Figure 5-5. Dose rates as a function of plutonium mass in assembly.

The calculations were repeated replacing the tantalum clad with molybdenum in order to determine if a reduction in parasitic absorption would reduce the assemblies strong dependence on neutron multiplication in achieving high dose rates (>100 rem/hr). Although the multiplication increased for smaller assembly sizes, the dose rates were similar for similar levels of neutron multiplication. Hence, the neutron multiplication was still a key factor in the RBA performance.

5.5 LEAKAGE ENHANCEMENTS

In an effort to reduce the dependence on high levels of neutron multiplication in achieving the desired dose rates, leakage enhancements were investigated. The first method for enhancing leakage that was tested was simply increasing the pitch-to-diameter ratio in the assembly. These calculations were performed for a 12X12 array of molybdenum/stainless-steel-clad pins of varying pin diameter. Figure 5-6 shows the neutron multiplication ($1/(1-k_{eff})$) for these cases. For 0.0175-m radii pins with a pitch-to-diameter ratio of 1.1, the assembly was critical and hence no multiplication is shown because it is infinite. For the remaining cases, it is evident from Fig. 5-6 that the multiplication increases with pin diameter (because the plutonium inventory is increasing) and decreases with pitch-to-diameter ratio (because leakage increases). Figure 5-7 shows the corresponding dose rates at 1 meter from the surface (at the axial mid-plane). As with the earlier calculations, the behavior of the dose rates closely mirrors that of the multiplication, indicating that the additional leakage is insufficient to offset the loss of neutron production through neutron multiplication.

The second method examined for enhancing leakage was by introducing an axial "flux trap". This "flux trap" is a void region in the axial direction which provides a large leakage path in the radial direction. It would be introduced into an RBA assembly by incorporating spacers

within the rods to separate groupings of the Pu-Be pellets by the desired gap; this is how it was explicitly modeled in the transport calculations. Single flux-traps located in the center of the assembly of various widths were introduced. The entire assembly was composed of a 16X16 array of 1-meter long tantalum/stainless-steel clad rods. Table 5-7 shows the results. A gap of zero (no flux trap) is included to allow easy comparison. As seen in Table 5-7, the k_{eff} decreases with increasing gap height. The dose rates also decreased correspondingly; the additional leakage provided by the flux trap was insufficient to offset the effects of the lower multiplication of the assembly.

Table 5-7. Results for axial flux-trap configuration for various gap widths.

Gap width	0 m	0.10 m	0.20 m	0.30 m	0.40 m
k_{eff}	0.839	0.756	0.700	0.645	0.588
Dose 1 meter from sides (rem/hr)	54	36	27	21	16
Dose 1 meter from top/bottom (rem/hr)	34	49	45	39	34

5.6 FLOODING CONCERNS

All of the options tested showed a close relationship between high dose rates and high levels of multiplication. Therefore, it is desirable to set the multiplication level of an RBA assembly as high as possible. The limitation on multiplication is set by safety concerns over the possibility of inadvertent criticality. This limit was set for RBA by maintaining a high degree of subcriticality (k_{eff}=0.95) under what was deemed the most credible scenario for a large positive reactivity insertion: flooding of the storage facility. Flooding was modeled by filling any open void regions with water and surrounding the assembly with 0.5 m of water on all sides. In addition, the outer surfaces of the water region were made reflective (i.e. no neutron leakage from the system) to ensure a conservative result. Initial calculations for RBA assemblies with the standard tantalum/stainless-steel cladding indicated that multiplication levels above k_{eff}=0.9 could not be used and still maintain the k_{eff}=0.95 limitation under flooded conditions. Several options were tested to alleviate the flooding effects and to maximize the allowable multiplication.

The first option tested as a method for reducing flooding effects was immersing the RBA array in a matrix of lead. In this manner, flooding between the rods is prevented, and the water can only surround the outside of the assembly. Lead was selected because its high atomic mass causes it to be a very poor neutron moderator, and its extremely low neutron absorption cross section keeps parasitic losses in the matrix material to a minimum. Lead also has the advantages of having a low melting point, allowing it to be melted and poured over the assembly with relative ease, being inexpensive (and available), providing an additional physical barrier to recovery of the plutonium, and providing an additional measure of structural integrity to the assembly (preventing any distortion of the rod geometry due to physical blows). Table 5-8 shows a comparison between a 16X16 array of tantalum/stainless-steel-clad rods in void and lead matrices. It can be seen that the lead actually increases the normal level of multiplication by decreasing neutron leakage. In addition, high external dose rates are still maintained, although they are less than a void matrix with the same level of neutron multiplication (not shown in Table 5-8). However, the lead does not prevent criticality under flooded conditions.

The second option investigated was the use of a thermal neutron absorbing material as a way to reduce the thermal neutron flux which is a small contributor to external dose, but a large factor in multiplication. Cadmium was selected because it has a high thermal neutron cross section (2450 barns), but a very low absorption cross section in the epithermal and fast energy regimes (the cross section drops off abruptly at 1.4 eV). Cadmium was introduced into the array in three separate ways: as a cladding material (replacing the stainless steel); as a matrix material

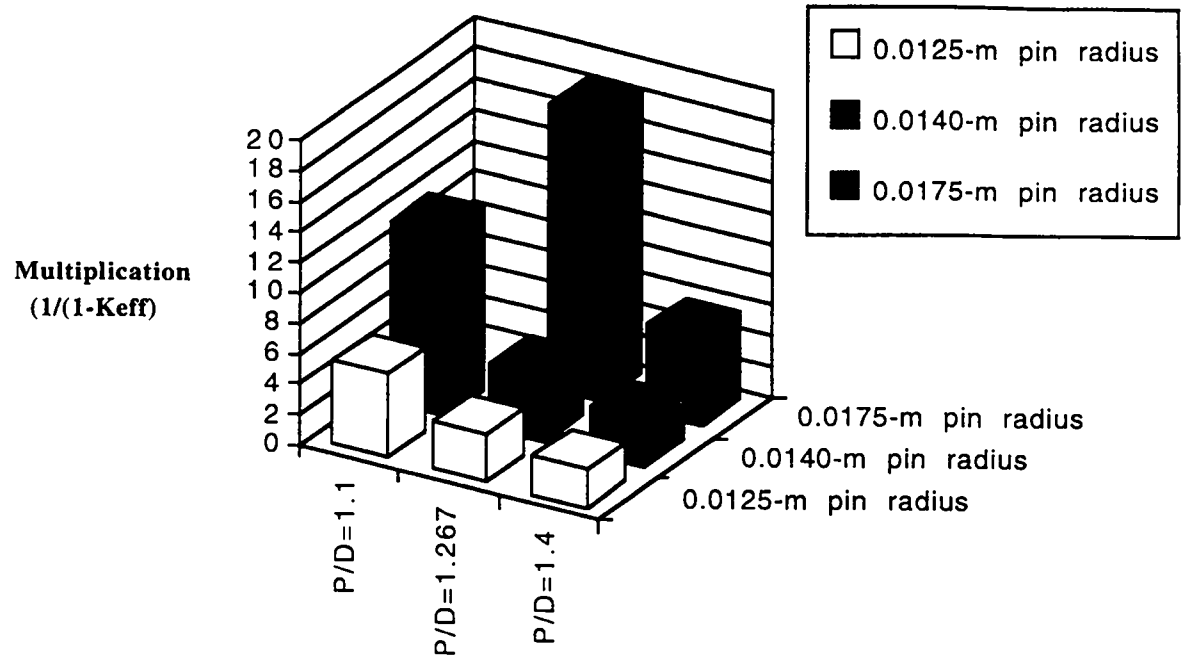

Figure 5-6. Neutron multiplication for various pitch-to-diameter ratios and pin radii (12X12 array of molybdenum/stainless-steel-clad pins).

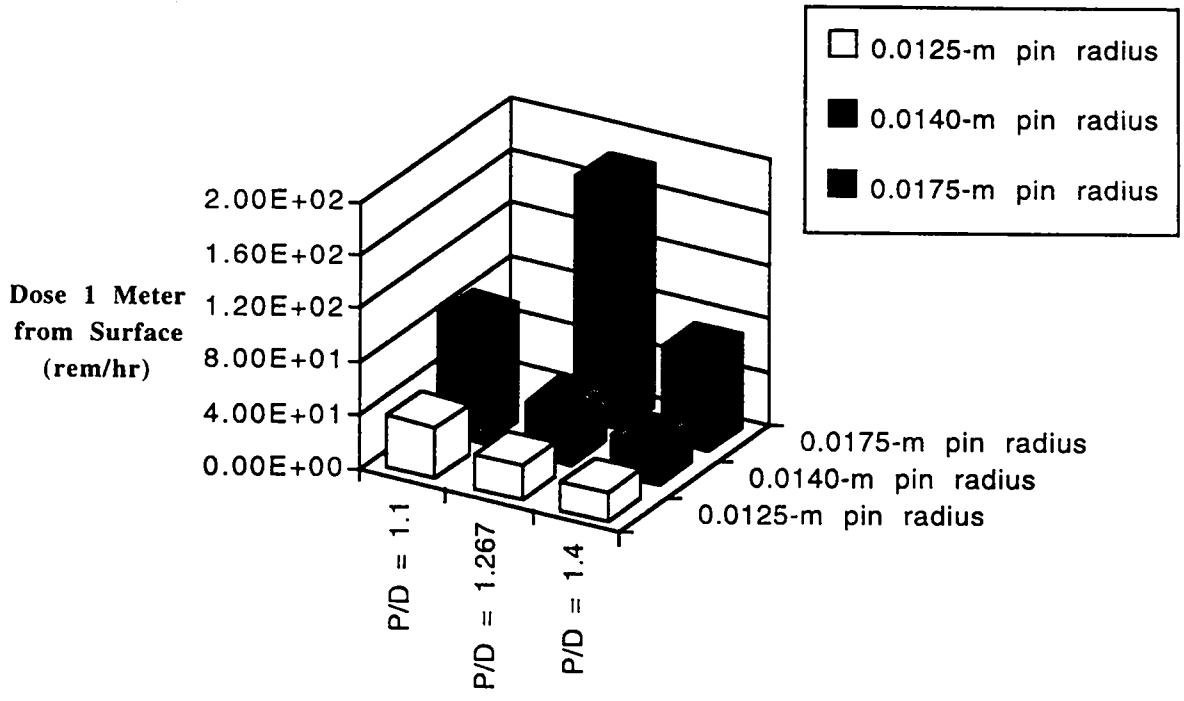

Figure 5-7. Dose rates for various pitch-to-diameter ratios and pin radii (12X12 array of molybdenum/stainless-steel-clad pins).

Table 5-8. Comparison of lead and void matrices for 16X16 tantalum/stainless-steel-clad arrays.

	Void Matrix	Lead Matrix
k_{eff}	0.84	0.96
k_{eff} (flooded)	0.94	1.02
Dose rate 1 meter from sides (rem/hr)	54	114
Dose rate 1 meter from top/bottom (rem/hr)	34	100

surrounding the pins; and as a can surrounding the entire array. The cadmium matrix had excessive self-shielding which produced extremely low external dose rates. However, both the cadmium clad and cadmium can prevented criticality under flooded conditions (for assemblies ranging in initial multiplication from k_{eff}= 0.90-0.95) without excessively reducing the external dose rates. The cadmium can produced the best results because less parasitic absorption occurred in the cadmium than in the clad case. Consequently, the use of cadmium in RBA assemblies (currently in the form of a can) was adopted for all subsequent RBA designs.

5.7 ASPECT RATIO VARIATION

Initial parametric calculations for RBA assembly configurations arbitrarily assumed a constant array length of 1 meter. To determine the optimum aspect ratio for the assembly as well as the effects of being off-optimum, a calculation was performed on a 16X16 tantalum/stainless-steel-clad array in which the aspect ratio of the overall assembly was varied. The resulting 1-meter dose rates are shown in Fig. 5-8. As expected, an aspect ratio of 1 results in an even distribution of dose among all sides of the assembly (which because a square pitch is being utilized is a cube).

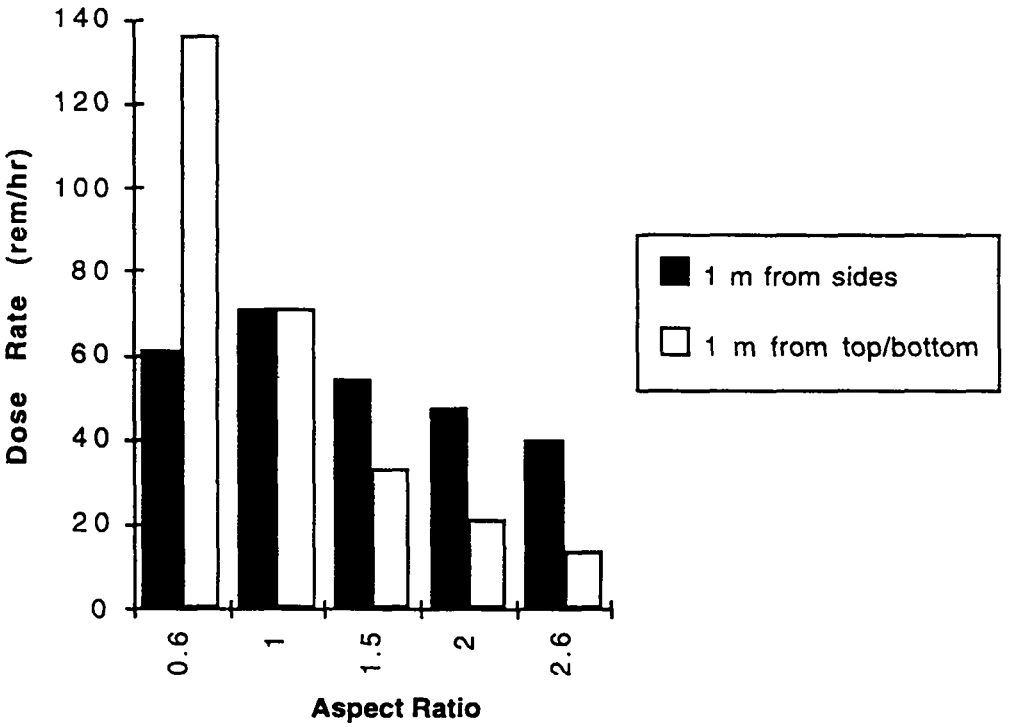

Figure 5-8. Effect of assembly aspect ratio on external dose distribution.

5.8 INCREASING EXTERNAL DOSE RATES

From the calculations outlined above, two main conclusions were reached: 1) neutron multiplication plays a key role in RBA performance; and 2) flooding concerns which arise from operating at high levels of multiplication can be reduced by the use of a thermal neutron absorber such as cadmium. However, the examined configurations generated marginal results (approximately at the 100 rem/hr level at 1 meter), even for high levels of multiplication. Consequently, efforts were initiated to increase the dose rates that were emitted.

To determine a method for increasing the dose rates, the factors which affect the dose were identified. For this purpose, the dose was represented as follows

$$\text{Dose rate} \propto \text{Source strength} \times \text{Neutron multiplication} \times \text{Dose conversion factor} \times \text{"}1/r^2\text{"} \quad (5\text{-}1)$$

where "$1/r^2$" represents the geometry effect on the dose value (this would be equal to $1/r^2$ if the neutron source was a point source). Of the factors in expression (5-1), the neutron multiplication is set by the maximum allowable k_{eff} for the RBA assembly and the dose conversion factor is maximized by minimizing the amount of moderation which occurs in the assembly (this maximizes the fast flux component which is the dominant dose contributor) which was always done in the examined configurations. To maximize the geometry factor (for the same total neutron source strength), the source should be concentrated into a point source. However, limits on the Pu-Be (α,n) source density and the need for significant multiplication constrain the size. The minimum-sized cylinder, achieved by using a pure Pu-Be source, has a diameter of ~0.38 m and a height of ~0.33 m and emits a nearly symmetric 1-meter dose rate of less than 40 rem/hr. Hence, even under ideal conditions, geometry manipulation is insufficient to increase the dose rates to the desired levels. Therefore, only the neutron source strength is available for manipulation in increasing the dose rates. Consequently, the following approach was adopted:

1) Set the neutron multiplication at the maximum allowable value. This value is assumed to be the multiplication of a bare assembly that when immersed in an infinite medium of water still meets the spent-fuel storage criteria ($k_{eff} \leq 0.95$).

2) Attempt to increase the neutron source strength to achieve the desired dose rates.

Two methods of increasing the neutron source strength were identified: 1) add shorter-lived alpha emitters (e.g., ^{238}Pu, ^{241}Am, etc.); or 2) increase the amount of Pu-Be in the assembly.

5.8.1 Addition of Shorter-Lived Alpha Emitters

The additional (α,n) source generation which can be obtained in RBA assemblies by adding ^{238}Pu or ^{241}Am was evaluated. Enhancement of the neutron source by the addition of these isotopes will result in an increase in the emitted dose. However, the final dose will depend on the multiplication of the assembly as well as the location of the isotopes within the assembly. Consequently, the only values reported here represent the increase in the initial (α,n) neutron source only. Design-specific calculations are required to determine any particular dose increases. Additionally, the time-dependent behavior of the dose would have to be evaluated and optimized with regard to the desired storage period because the neutron source will decay away faster than for ^{239}Pu.

The neutron yield emitted from beryllium per alpha particle as a function of alpha energy was obtained from reference (8) and is plotted in Fig. 5-9. The data was fit with a third-degree least-squares polynomial (which is shown in Fig. 5-9) as

$$Y\ (n/alpha) = -5.8442\times10^{-7}E^3 + 1.6651\times10^{-5}E^2 - 8.3971\times10^{-5}E + 1.2743\times10^{-4} \quad (5\text{-}2)$$

where Y is the neutron yield in neutron per alpha and E is the alpha energy in MeV. Table 5-9 shows the dominant alpha energies emitted by decay of ^{238}Pu, ^{239}Pu, and ^{241}Am as well as the average alpha energy of each isotope. The average alpha energies shown in Table 5-9 were used in conjunction with equation (5-2) and the decay characteristics shown in Table 5-10 of the isotopes to predict the relative source strengths. A summary of these values is given in Table 5-11. Using the values in Table 5-11, plots were generated for the source enhancements obtained for various additions of ^{238}Pu and ^{241}Am. These are shown in Figs. 5-10 and 5-11, respectively.

It can be seen from Fig. 5-10 that the addition of ^{238}Pu provides a significant enhancement in neutron source even for replacement of less than 1% of the ^{239}Pu. Figure 5-11 shows that the addition of ^{241}Am can also enhance the source, but significant gains are not obtained unless at least a few percent of the ^{239}Pu is replaced. Altogether, addition of either or both of these isotopes appears to be an attractive way of sizably increasing the neutron source, and hence the emitted dose, of an RBA assembly. However, a review of the availability of these isotopes in the DOE complex indicates that insufficient amounts are available for use in RBA.

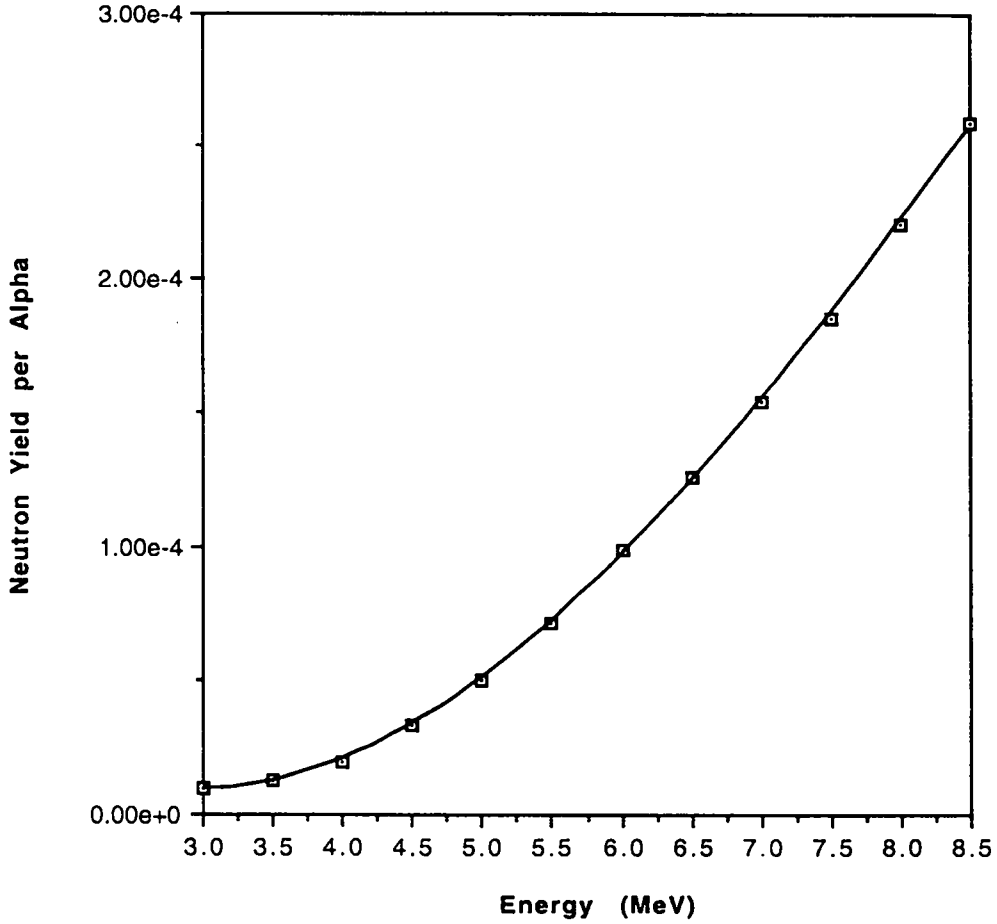

Figure 5-9. Neutron yield from Pu-Be (α,n) reactions.[8]

Table 5-9. Dominant alpha-decay characteristics[9] for ^{238}Pu, ^{239}Pu, and ^{241}Am.

^{238}Pu	
Alpha Energy (MeV)	Fraction of Decay
5.4980	0.711
5.4540	0.287
5.3590	0.002
Average Alpha Decay Energy (MeV)	5.4851

^{239}Pu	
Alpha Energy (MeV)	Fraction of Decay
5.1554	0.734
5.1429	0.151
5.1046	0.115
Average Alpha Decay Energy (MeV)	5.1477

^{241}Am	
Alpha Energy (MeV)	Fraction of Decay
5.486	0.8498
5.443	0.136
5.389	0.0142
Average Alpha Decay Energy (MeV)	5.4788

Table 5-10. Decay characteristics[10] used to compute relative source strengths.

Isotope	Half-life (years)
^{238}Pu	87.74
^{239}Pu	24110
^{241}Am	432

Table 5-11. Relative (α,n) source strengths calculated using Equation (5-2) and decay characteristics from Tables 5-9 and 5-10.

Isotope	Relative Source Strength
^{238}Pu	346
^{239}Pu	1
^{241}Am	70

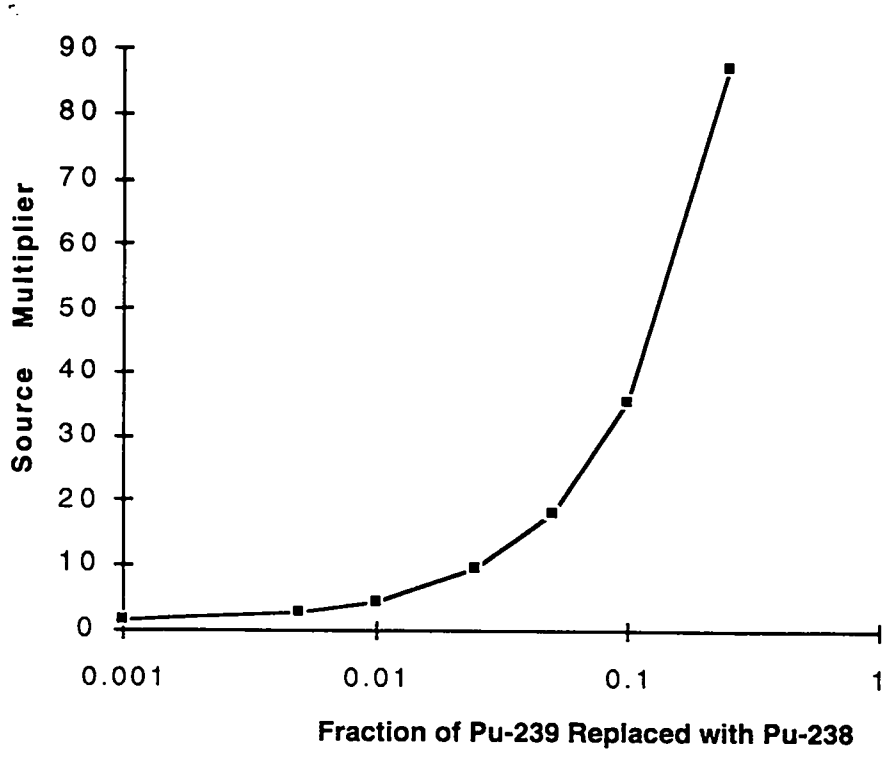

Figure 5-10. Neutron source enhancement by ^{238}Pu addition.

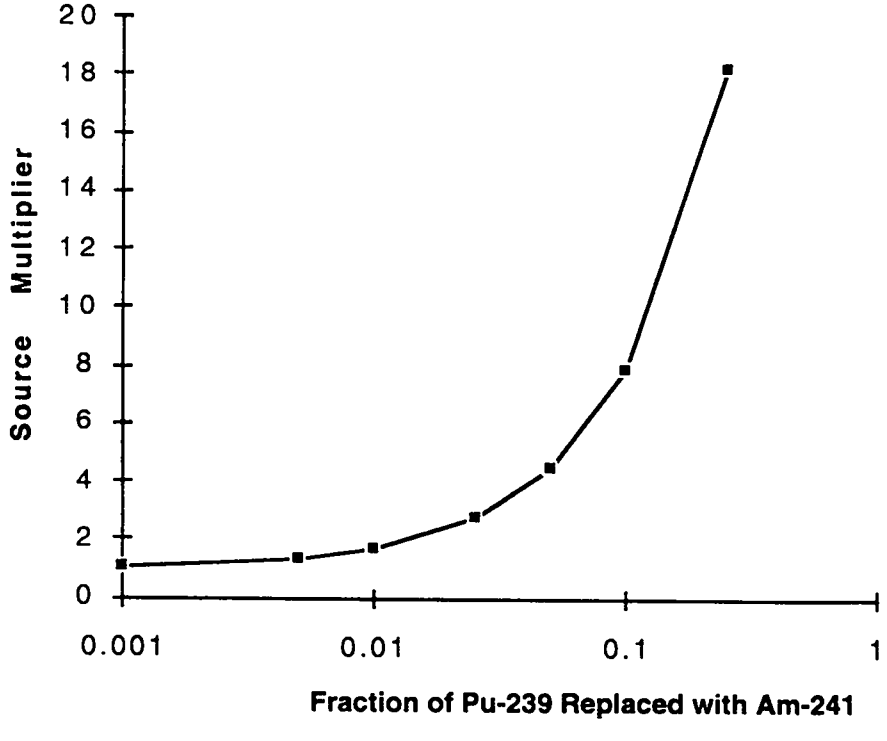

Figure 5-11. Neutron source enhancement by ^{241}Am addition.

5.8.2 Increasing the Amount of Pu-Be

While increasing the amount of Pu-Be in an RBA assembly results in a proportional increase in the (α,n) source strength, it also causes an increase in multiplication unless the additional neutron production through fission is offset either through absorption or leakage. Since the goal is to generate high dose rates external to the assembly, leakage was the selected mechanism. As demonstrated in Section 5.5, various methods exist for increasing the neutron leakage. However, in an effort to maintain a uniform dose distribution around the entire RBA assembly, the leakage was increased by increasing the rod pitch-to-diameter ratio. Therefore, the array was increased in size to give dose rates of at least 450 rem/hr at 1 meter from the surfaces of the assembly, and then the pitch-to-diameter ratio was increase until a k_{eff} of 0.95 under flooded conditions was obtained. This value assumes encasing the entire array in a 0.01-m thick cadmium can to minimize the flooding effects. The k_{eff} for the bare assembly was 0.91. The resulting dose rates ranged from 540-640 rem/hr at 1 meter, and consequently this was selected as the baseline RBA configuration for further engineering analyses.

Although increasing the amount of Pu-Be in an RBA assembly is successful in generating the desired dose levels, it has some additional consequences. The inventory of plutonium in each assembly becomes very large (~ 9 MT), and the entire mass of the configuration makes transportation of the full assembly prohibitively difficult (although the assembly could constructed of smaller assemblies which might be suitable for transportation); thus the assembly will have to be constructed to some degree in its final storage location. The large size also has the potential to cause a high centerline temperature if the assembly is encased in a matrix (lead) which would rely solely on conduction for internal heat removal. The effects of these issues must still be evaluated.

5.9 RBA BASELINE STORAGE CONCEPT

The characteristics of the RBA baseline storage concept are given in Table 5-12. This configuration will be utilized for ongoing engineering analyses to complete the feasibility study for the RBA concept.

Table 5-12. Characteristics of RBA baseline storage concept.

Array Size (Number of Pins)	36 X 36
Total Number of Pins	1296
Pu-Be Diameter (m)	0.032
Inner Clad (Tantalum) Thickness (m)	0.0025
Outer Clad (Stainless Steel) Thickness (m)	0.0025
Pitch (m)	0.09
Pitch-to-Diameter Ratio	2.14
Outer Can (Cadmium) Thickness (m)	0.01
Assembly Dimensions (m)	3.21 X 3.21 X 3.02
Assembly Volume (m^3)	31
Total Assembly Mass (kg)	45,900
Total Pu-Be Mass (kg)	13,600
Total Plutonium Mass (kg)	9,100
Total Tantalum Mass (kg)	17,500
Total Stainless Steel Mass (kg)	9,700
Total Cadmium Mass (kg)	5,200
Total (α,n) Neutron Source (n/s)	9.11e+11
K_{eff}	0.91
K_{eff} (flooded)	0.95
Dose 1 Meter from Side (rem/hr)	536
Dose 1 Meter from Top (rem/hr)	640

6.0 ENGINEERING ANALYSES

Detailed engineering analyses must still be performed on the RBA storage concept. The following two sections (Sections 6.1 and 6.2) briefly examine two major engineering issues: heat removal from the assemblies, and gas generation. However, a number of additional engineering issues exist (*e.g.*, assembly/disassembly, spacer design, welding of materials, *etc.*) which must be addressed for the final concept evaluation.

6.1 HEAT GENERATION AND REMOVAL

Because approximately 17,000 decays must occur per neutron generated through the (α,n) process, the main form of energy deposition in RBA is through decay. This process produces a power density of 6.68e-03 W/cm^3 in the Pu-Be regions. In comparison, energy produced through fission, other neutron interactions, or gammas interactions results in an additional power deposition of 1.56e-05 W/cm^3. For the baseline storage concept, a total of 21 kW is generated in an RBA assembly. Because of the very low power densities and amount of power generated, heat removal is not considered a severe problem. However, full three-dimensional heat transfer models are currently being developed to predict the temperature distribution within RBA rods and for the entire RBA assembly. For this purpose, the IDEAS software package is being used. This package has the capability of modeling conduction, convection, radiation, advection, phase change, and fluid flow, as well as structural analysis. Of possible concern is heating in large arrays with a surrounding lead matrix which could lead to unacceptable high temperatures in the center of the assembly. This will soon be evaluated and any problems identified.

6.2 GAS PRODUCTION

Because of the large amount of decays per neutron generated, the helium produced through alpha-decay is the dominant gas produced in RBA assemblies (as opposed to gaseous fission products). The gas production will have to be accommodated for by the incorporation of a gas plenum within the inner cladding of each RBA pellet. The gas buildup will result in an increase in stress on the cladding material. Models are being developed using the IDEAS software package mentioned above to quantify these stress levels, and to predict cladding lifetimes based on the stress to which they are exposed.

7.0 EFFECTS ON STORAGE FACILITY DESIGN

The effects of RBA implementation on the storage facility design remain to be evaluated. Obviously, the use of RBA incurs the requirement for remote handling at the storage facility. In addition, the coupling of the storage facility to the RBA fabrication facility, including the effects of the location of each and any required transportation, also must be evaluated. For the baseline RBA configuration, approximately 6 RBA assemblies, each 31 m^3 in volume, would be required to store 50 MT of weapons-grade plutonium. A schematic representation of this is shown in Fig. 7-1. This configuration results in a much smaller storage volume requirement than other storage options (such as storing as pits). The cost savings due to this, and the cost penalties incurred in the RBA fabrication and remote handling requirements must be evaluated in order that a full cost-benefit analysis can be completed for the use of RBA.

8.0 CONCLUSIONS

The Radiation Barrier Alloy (RBA) concept appears to be an attractive method for introducing radioactive, chemical, and physical barriers for use in storage of weapons-grade plutonium, and yet still allow for accurate material control and accountability, as well as for retrieval of the material by the host nation if desired. A preliminary feasibility study has been initiated, and the following conclusions have resulted:

- Dose levels in excess of 500 rem/hr at a 1-meter distance from the surface of the RBA assembly can be obtained.
- Essential for achieving these dose levels is operation at a high level of neutron multiplication ($k_{eff} \sim 0.9$).
- Criticality concerns, even under flooded conditions, can be eliminated through the use of a thermal-neutron-absorbing material (*e.g.*, cadmium) either as a cladding material or a container material surrounding the RBA assembly.
- Fabrication techniques for the Pu-Be compound are well demonstrated and fully compatible with the RBA assembly fabrication.
- Data from disassembly of Pu-Be sources indicate that the compound is stable and no significant physical degradation occurs over a 40-year time frame. There is no reason to believe that any additional problems exist for longer time frames, given that the components are designed for the appropriate lifetimes (*i.e.*, adequately account for gas production).
- The materials required for RBA implementation are available in the required quantities. Cost of these materials is not prohibitive. The possible exception is tantalum (see Section 4.1), although its use is non-essential for RBA performance and hence it will probably be eliminated from future RBA designs.
- Additional physical barriers can be added by welding the assembly together, and encasing the assembly in an outer container. If desired, the assembly (inside the outer container) can also be immersed in a neutronically-inert matrix such as lead.

To further the RBA preconceptual analyses, a baseline design based on physics performance was developed. Based on this design, detailed thermal-hydraulic and structural stress calculations are being performed. In addition, an evaluation of the effects on the storage facility and a cost-benefit analysis for the RBA approach are being conducted. For the baseline RBA configuration, approximately six RBA assemblies, each 31 m^3 in volume, would be required to store 50 MT of weapons-grade plutonium. A schematic representation of this is shown in Fig. 7-1. This configuration results in a much smaller storage volume requirement than other storage options (such as storing as pits). Completion of these analyses will allow a better evaluation of the benefits of the use of RBA in the plutonium disposition process.

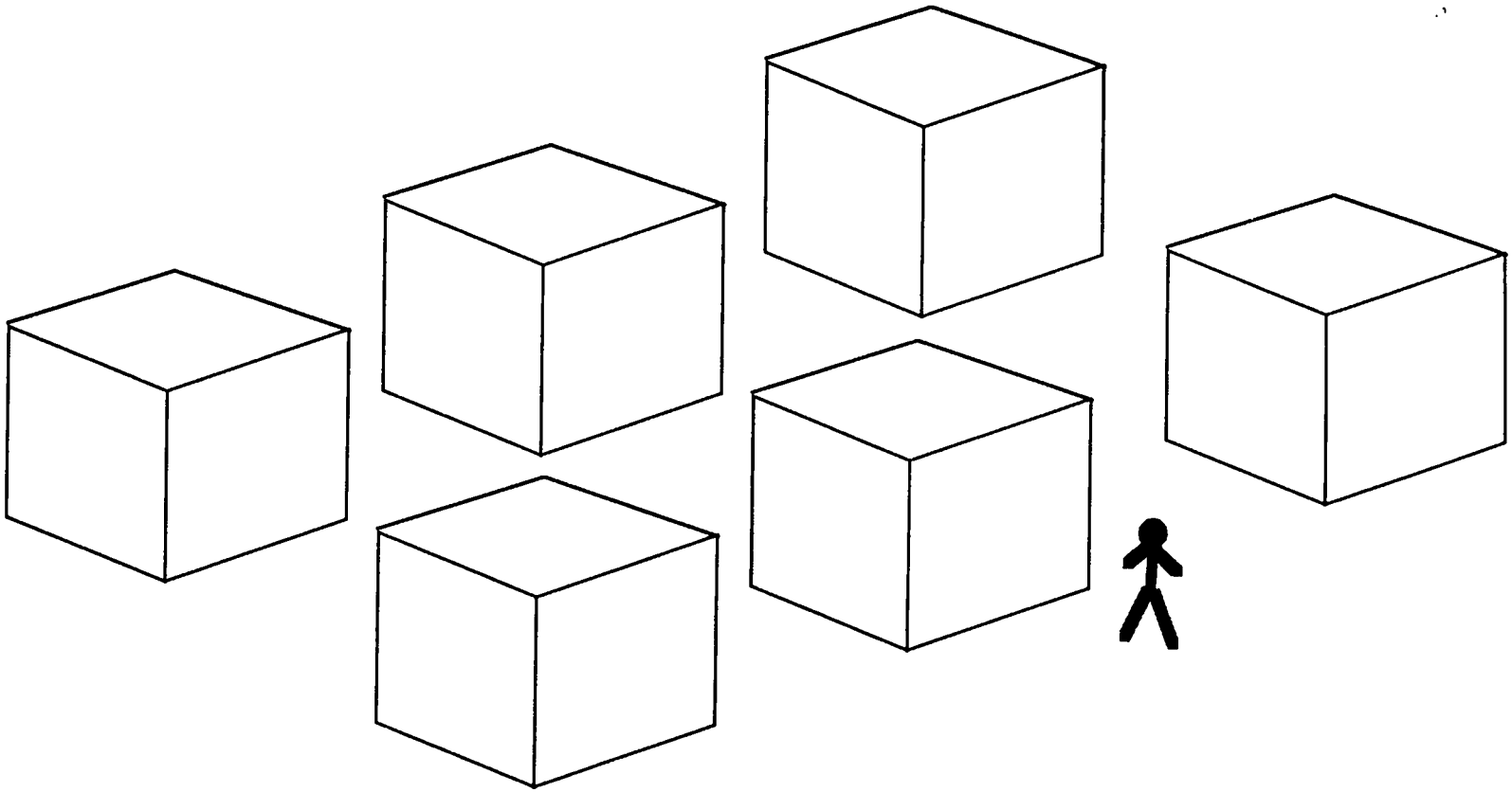

Figure 7-1. Schematic representation of RBA storage.

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