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Advanced Concept Proof-of-principle Demonstration: Switchable Radioactive Neutron Source

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ADVANCED CONCEPT
PROOF-OF-PRINCIPLE DEMONSTRATION:
SWITCHABLE RADIOACTIVE NEUTRON SOURCE

By

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ADVANCED CONCEPT
PROOF-OF-PRINCIPLE DEMONSTRATION:
SWITCHABLE RADIOACTIVE NEUTRON SOURCE

E. A. Rhodes, D. L. Bowers, R. E. Boyar, and C. E. Dickerman

Executive Summary

An advanced concept proof-of-principle demonstration was successfully performed to show the feasibility of a practical switchable radioactive neutron source (SRNS) that can be switched on and off like an accelerator, but without requiring accelerator equipment such as high voltage supply, control unit, etc. This source concept would provide a highly portable neutron source for field radiation measurement applications. Such a source would require minimal, if any, shielding when not in use. The SRNS, previously patented by Argonne staff, provides a means of constructing the alpha-emitting and light-element components of a radioactive neutron source, in such a fashion that these two components can be brought together to turn the source "on" and then be separated to turn the source "off".(1) An SRNS could be used for such field applications as active neutron interrogation of objects to detect fissile materials or to measure their concentration; and to excite gamma-ray emission for detection of specific elements that indicate toxic chemicals, drugs, explosives, etc.

The demonstration was performed using Pu-238 as the alpha emitter and Be as the light element, in an air-atmosphere glovebox having no atmosphere purification capability. A stable, thin film of Pu-238 oxide was deposited on a stainless steel planchet. This deposition process was selected because it did not require significant development work. The "on" output of the demonstration Pu-238 film was measured to be 2.5×10^6 neutrons/sec-gram of Pu-238, in agreement with the estimate made during development of the concept. The measured "off"neutron rate was satisfactory, only about 5% of the "on" output, after two weeks of exposure to the glovebox atmosphere. After several weeks additional exposure, the "off"rate had increased to about 15%. This result indicates that short-term operations on the plated surfaces for actual SRNS units would not require special atmospheric control, but that preparation, storing, and assembly operations should be carried out in a high-purity atmosphere. This work demonstrates the feasibility of constructing practical, highly portable SRNS units with very low gamma-ray dose in the "off" position.

1.0 INTRODUCTION

The switchable radioactive neutron source (SRNS) is an ANL concept covered by U.S. Patent 4,829,191.(1) This device is a highly portable neutron source that can be switched on and off like an accelerator, but without requiring accelerator equipment such as high voltage supply, control unit, etc. Thus, the SRNS requires little, if any, neutron shielding for transportation or personnel protection when not in use (unlike a conventional radioactive neutron source). The SRNS emits no significant gamma-ray background when not in use (unlike a Cf-252 fission source). The reference concept design emits 10^8 neutrons/sec, and contains approximately 40 grams of Pu-238. This corresponds to a specific neutron output of 2.5×10^6 neutrons/sec-gram. The mechanical design permits accurate and reproducible "on" and "off" settings of the Pu-238 and Be components. An SRNS could be used for such field applications as active neutron interrogation of objects to detect fissile materials or to monitor their concentration; and to excite gamma-ray emission for detection of specific elements that indicate toxic chemicals, drugs, explosives, etc.

Although this SRNS was a technically attractive concept, significant uncertainties existed on the practical implementation of the concept. These concerns included: The ability to obtain a stable Pu-238 film; the ability of a sufficient fraction of alphas to escape from the film to generate an adequate neutron "on" output from this film, and the background neutron emission "off" rate (from light element atoms in and on the film). Because of the strong energy absorption of alphas, the film thickness had to be of the order of only ten microns or less, in order to assure escape of a significant fraction of the alpha emission. Accordingly, a small Advanced Concepts project was set up to provide a proof-of-principle demonstration of the concept. This report documents the successful completion of the demonstration.

The demonstration film was prepared using a chemical deposition process that deposited a film, of about three microns average thickness, consisting of Pu-238 in the stable oxide form, on a standard stainless steel planchet. (Other approaches, such as sputtering of Pu metal, should provide films with higher performance, but at the expense of significant development work well beyond the scope of this small demonstration project.)

2.0 SWITCHABLE RADIOACTIVE NEUTRON SOURCE

2.1 Concept

Figure 2-1 is a drawing of the SRNS patent configuration. The heart of the mechanical design is the parallel plates that contain surfaces of the alpha-emitting film and the light element "target" which emits neutrons when exposed to alphas. This arrangement provides for separation of the alpha-emitting nuclei from the light nuclei that emit neutrons via the (alpha,n) reaction, and it is the key feature that provides this concept with the unique ability to switch the radioactive neutron output "on" and "off". Each plate is divided into segments that match those of the adjacent plates; alternate plates are in a fixed position.

1. Double-walled capsule
2. Rotatable shaft
3. Bearing
4. Armature driven by external electromagnet
5. Bearings
6. Circular alpha-emitting disk

FIG. 1

7. Circular neutron-emitting disk
8. Shaft key
9. Alignment key
13. Bracket
14. Rotary solenoid to drive electromagnet
15. External electromagnet
16. Alignment coil spring

Fig. 2-1. SRNS patent diagram.

Those plates between the fixed plates can be rotated into the "on" position so that the alpha-emitting surfaces are directly opposite the light-element surfaces and alphas can strike the light element, or alternately, into the "off" position in which alphas from the alpha-emitting surfaces cannot strike the light element.

The concept illustration of Fig. 2-1 has a non-magnetic stainless steel capsule, with a magnet-actuated mechanism for rotating the movable plates. No moving seals are required to move the plates. The capsule can be evacuated, or alternatively, can contain a non-chemically contaminating atmosphere.

Figure 2-2 illustrates this SRNS "on"- "off" operation. The plate sectors coated with Pu-238 are indicated by shading. Sectors with surface Be are unshaded. For clarity, the figure shows only two adjacent plates, separated sufficiently to expose the relative orientation of beryllium and Pu-238 deposits on adjacent plates. In an actual system, these plates would be much closer together, to minimize the probability that an alpha emitted at a small angle with respect to the surface could strike the wrong sector in the opposite plate, and there also might be small baffles to intercept and absorb such low-angle alphas. On the left side of the figure are two plates in the "off" position. The alpha-emitting deposit "sees" only another alpha-emitting deposit. On the right side of the figure these two plates are rotated into the "on" position, and alphas from each alpha-emitting deposit strike only beryllium, with the emission of (alpha,n) neutrons.

A nonmagnetic stainless steel would probably be a suitable material for the disks and capsule--for rotor control, structural strength, low neutron background, and relatively low activation. The depths of the alpha-emitter and target layers on the disks need not be much larger than the respective alpha-particle ranges, as no further increase in neutron yield is provided by thicker layers. A thin submicron coating over the alpha-emitter layer would prevent flaking and emission of recoil nuclei, to avoid buildup of neutron background in the "off" position. Disk diameter and numbers of disks would depend on the source container size and neutron source strength requirements for specific applications. An electromagnetic switching mechanism is indicated.

The capsule concept shown in Fig. 2-1 could easily be carried in the field by one person. Four time-coded operational modes have been identified to provide a range of potential applications: steady-state (on-off), intermittent pulses, slow pulsing (up to ten revolutions per second), and fast pulsing up to hundreds of revolutions per second. Simple on-off hand operation would be suitable for steady-state applications.

2.1.1 Suitable Targets for (alpha,n) Reactions

The light elements used as target material on the disks should be chosen for relatively high yield of (alpha,n) neutrons in the energy range of interest, leading to the choice of Be, B, and Li for our initial calculations. Properties of these elements as target elements, when deposited as a layer with thickness nominally larger than the range of an alpha particle of about 6 MeV, are shown in Table 2-1. Be provides the highest idealized neutron yield and highest neutron energies, B is intermediate in yield and energy, and Li gives relatively low yield and low energy. These idealized yield values assume no alpha attenuation

Fig. 2-2. Schematic of "on-off" operation.

or energy degradation in the emitter layer. As shown in the table, gamma rays can be emitted as well as neutrons when alphas from the emitter strike the target [this, of course, occurs for either conventional (alpha,n) sources or a SRNS]. Be has the highest gamma-ray output per neutron, B is next, and Li is lowest. Although the ranges of relative neutron yields shown in the table would be expected to apply for a range of alpha emitters, the absolute yields depend strongly upon the alpha energies of the specific emitter. Relatively wide ranges in potential neutron output are shown in the table, because the three target materials have distinctly different yields as a function of alpha energy. The next to last row of Table 2-1 presents total yield calculated, after taking into account alpha attenuation and energy losses in the emitter. The calculations are for emitter and target layer thicknesses nominally larger than the range of alphas having the initial energies, and depend more on the target material than on the emitter. There is a relatively small correction depending on the energies of the alphas emitted. We estimate that only about 25% of the alpha particles emitted within the emitter reach the adjacent target layer. These undergo substantial energy losses before reaching the target, resulting in substantially reduced neutron yield. We also assume a small energy loss due to the emitter coating (assumed to be equivalent to 0.17 micron of Pd).

Table 2-1. Properties of selected target materials

Target	Be	B	Li
Average N Energy, MeV	~4.5	~2.7	~0.5
Max. N Energy, MeV	~12	~5	~1.5
Idealized Neutron Yield, no transmission losses, % of Be	100	~25-50	~0.8-5
Gamma Energy, Mev	4.43	2.31	negl.
Gammas per Neutron	~0.8	~0.07	negl.
Estimated Neutron Yield, with transmission losses, % of Ideal	~7	~10	~3
relative to Be, %	100	~36-71	~0.3-2.1

The yields from these alphas were approximated from computations based on thick-target yield data, fitted to the empirical formula:

$$Y = b E^v,$$

where Y is yield, E is energy, and b and v are empirical constants. In addition, it was assumed that alpha range is proportional to alpha energy raised to the 3/2 power, there is no range or energy straggling, and the emitter layer is uniform. The resulting values are expressed as a percentage of the yield from the idealized case of no alpha attenuation or energy losses in the emitter. The last row of the table translates these yields into values relative to the idealized output for the Be target, for convenience in comparison. The data of Table 2-1 indicate that one might choose Be when yield and high energy are important,

B when an approximation to the fission spectrum is important, and Li when easily thermalized neutrons and absence of high-energy neutrons are important.

2.1.2 Possible Alpha Emitters

Suitable alpha-emitting isotopes for a SRNS will have the following properties: (a) a half-life sufficiently long for the source to have a useful lifetime, but sufficiently short to produce a useful output, (b) sufficiently small spontaneous neutron output when the source is switched "off" compared with the neutron emission when the source is switched "on", (c) acceptably small spontaneous gamma-ray background, (d) daughter products that contribute sufficiently small background radiation, and (e) availability in gram quantities at an acceptable cost. If the emitter film consists of, or contains a compound including light elements (C, O, N, etc.) it will have a spontaneous background contribution from (alpha,n) reactions that cannot be switched "off".

Table 2-2. Properties of selected (alpha,n) sources

Alpha Emitter		Th-228	Cm-242	Po-210	Ac-227	Pu-238	Am-241
Half-life		1.91 y	163 d	138 d	21.8 y	87.8 y	432 y
Gamma-ray Energy, keV		80-2,600	44	800	50-870	44-766	60
Gamma Dose at 1 meter, mRem/hr-cm ²		17,000	0.73	0.45	230	0.003	0.63
Neutrons per cm ² -sec.	Be	2,100	1,900	1,200	190	5.9	1.1
"on", x 10 ⁴	B	600	610	470	55	2.2	0.4
	Li	100	10	10	8.4	0.064	0.012
Neutron "On-Off" Ratio	Be	high	41	high	high	1,250	high
	B	high	14	high	high	460	high
	Li	high	2	high	high	15	high

Results of a survey of six candidate isotopes are shown in Table 2-2 listed in order of decreasing neutron yield. Calculations assumed that the alpha-emitter layer thickness is equal to the range of the highest energy alpha of the emitting isotope, generally equivalent to about 20 mg/cm². The gamma-ray doses were calculated at one meter from each emitter per square cm of emitter layer, and do not include any gamma-ray dose from (alpha,n) reactions. The neutron emission for each "on" case also was calculated per square cm of emitter, using the approximate calculation techniques described in Section 2.1.1 above, and do not include losses due to edge effects at disk layer boundaries, or neutron attenuation in construction materials. Neutron emission for each "off" case is the spontaneous

neutron emission from the pure metal per square cm of emitter. The nuclides Th-228 and Ac-227 were assumed to be in equilibrium with their daughter products. The alpha emission from these daughters enhances the neutron emission from the respective targets, but the daughters also give relatively high doses of penetrating gamma-rays that cannot be switched off. However, as will be discussed later, (alpha,n) reactions from other elements present in the emitter layer (and possibly, spontaneous fissions from heavy-metal impurities) may be the dominant source of "off" neutrons. Thus this table may be considered to provide upper limit estimates of the "on-off" ratios attainable from the specific isotopes listed.

The high gamma-ray emission from Th-228 and Ac-227 led to their elimination from consideration, because of the desire to avoid a SRNS that requires heavy gamma-ray shielding. Cm-242 and Po-210 were eliminated because of relatively short half-lives. These two isotopes could be suitable for special applications requiring high output over time spans compatible with their half-lives. Of the remaining two isotopes, Pu-238 was selected over Am-241 because Pu-238 has a projected neutron output per unit surface area of emitter of about five times that of Am-241. Of the three light target elements considered, Be had about three times the projected neutron output of B, and about one hundred times the projected output of Li. For these reasons, the material couple selected for this SRNS Advanced Concepts demonstration was Pu-238 and Be.

Plutonium oxidizes readily even in relatively high-purity inert gas atmospheres. It was our judgement that a process based on a metallic film would require a development effort requiring significantly larger resources than those available for this demonstration. Thus, the design approach selected for this demonstration of a practical SRNS uses a film of Pu oxide, in order to provide a stable deposit. We recognized that this approach automatically incorporates natural oxygen inside the film, ensuring that there is a built-in background of neutrons produced in the film from the (alpha,n) reaction with natural oxygen. The specific film deposition technique used for the actual demonstration also included application of a very thin collodion layer to help stabilize the film. This layer provided additional O and C atoms. Scoping estimates, presented in Section 2.2, indicated that the "off" neutron emission from this built-in background would be acceptable; but clearly, a demonstration was needed. (Alternate approaches to achieving a film with much lower "off" neutron output are identified in Section 4.0.)

2.2 Projected Performance

2.2.1 Estimated Neutron Output in the "On" Position

Pu-238 has a 88-yr half-life. The alpha yield is 6.4×10^{11} alphas/sec-gram.(2) From Table 2-2, a neutron output of 5.9×10^4 neutrons/sec-cm² is obtained for a pure metallic Pu-238 emitter coupled with a Be target. The calculations for this case assumed a uniform deposition of 18.2 mg/cm² of Pu-238 metal, leading to an estimate of 3.24×10^6 neutrons/sec-gram for neutron output in the "on" position. Estimating a further 12% loss due to edge effects caused by the finite sizes of the emitter film and target we obtain 2.82×10^6 neutrons/sec-gram. This is an upper-bound estimate. Film coating, non-uniformity of the film, and the presence of oxygen in the emitter film will reduce the output from a real system to a lower value. Note that this estimate is less than 10% of that obtained from a

simple calculation using the Pu-238 alpha output and thick target (alpha,n) data, reflecting the fact that these corrections are significant in a practical system.

2.2.2 Estimated "Off" Neutron Background Output

The spontaneous fission neutron yield of Pu-238 is 2.59×10^3 neutrons/sec-gram.(3) If the Pu-238 film were to contain no light nuclei such as Be, O or N, then the "off"neutron emission of the SRNS would result only from this spontaneous fission neutron source, which is about 0.1% of the projected "on" output given in Section 2.2.1.

However, if these alpha-active nuclei are contained in a film with light nuclei capable of emitting neutrons by the (alpha,n) process, this film will emit some (alpha,n) neutrons, even when the system is in the "off" configuration. The deposition process used in the demonstration deposits Pu in the oxide form; further, a thin (< 200 microgram) coating of collodion was deposited on the film to stabilize the film, as part of the post-deposition film processing. And, finally, post-deposition surface contamination might occur in the glovebox used for processing, storing, and assembly of a SRNS. Thus, concerns were raised about the background neutron emission from a practical SRNS deposit, resulting from light element presence in/on the Pu-238 film. This is why it was considered to be necessary to obtain experimental data on both the "on" and the "off" neutron emission from a practical film, under conservative conditions of exposure to glovebox atmosphere.

We may estimate the order of magnitude of "off"neutron emission from the oxide film using the datum of 1.34×10^4 neutrons/sec-gram emitted from the (alpha,n) reaction in Pu-238 oxide.(2) This value applies to (alpha,n) emission from bulk oxide; and it should provide an over-estimate of neutron production within an oxide film that is thin enough to allow a significant fraction of alphas to escape. The neutron emission calculated using this process is significantly larger than the spontaneous fission neutron emission; however, it is only about 0.5% of the estimated "on" neutron emission from this material couple.

We consider the total "off"neutron emission fraction of 0.6% of the "on" position neutron output estimated above by summing the outputs calculated for spontaneous fission and (alpha,) reactions in the oxide to be a lower-limit estimate for "off"neutron emission, because of uncertainties including the effects of non-uniform film deposition, and the possibility of significant quantities of local contamination.

2.2.3 Gamma-ray Background in the "Off" Position

The isotopic source combination of Pu-238 and Be is recognized as a very low gamma-emission arrangement. For example, Ref. 4 quotes a dose of 0.006 mRem/hr at a distance of one meter from a typical Pu-238-Be source emitting 1×10^6 neutrons/second.

We estimate the generic gamma-ray dose to personnel handling the SRNS in the "off" position, from the Pu-238 using the spontaneous fission gamma-ray output and the dose from the 0.766 MeV gamma ray. (The other gamma rays emitted in conjunction with the alpha decays are neglected in estimating gamma-ray dose, because they are of low energy, with low transmission through the internal structure and wall of a practical SRNS capsule.)

Pu-238 has an average value of 2.21 neutrons emitted per fission, and a spontaneous fission neutron yield of 2.6×10^3 neutrons/sec-gram.(3) Using an average gamma-ray emission (prompt plus delayed) of about 16 MeV per fission, we predict a spontaneous fission gamma-ray energy yield of 1.9×10^4 MeV/sec-gram. The emission rate for 0.766 MeV gamma rays is 1.39×10^5 gammas/sec-gram.(5) The total gamma-ray energy output is thus 1.25×10^5 MeV/sec-gram. Using this gamma-ray energy output, and an approximate conversion factor of 6×10^6 MeV/cm²-sec = 1 Rem/hr, the fission gamma-ray dose rate at a distance of only 20 cm from a point source of one gram of Pu-238 is calculated to be about 0.02 mRem/hr. Thus, the gamma-ray dose from this switchable source in the "off" position may be neglected.

2.2.4 Gamma-ray Background in the "On" Position

Table 2-2 indicates the 4.43 MeV gamma-ray emission from the (alpha,n) reaction on Be to be about 0.003 mRem/hr for 18.2 mg Pu-238 at a distance of one meter. For one gram of material, this becomes 0.16 mRem/hr at a distance of one meter.

2.3 Comment on Nuclear Material Safeguards

Under the U.S. Department of Energy Graded Safeguards Program, Pu-238 with isotopic composition greater than 60% is "Attractiveness Level E: All Other Nuclear Materials".(6)

Under DOE Order 5633.3B, Attractiveness Level E materials are to be treated under Category IV requirements. Category IV is the lowest Category; for example, depleted uranium falls under Category IV. Category III is the next higher Category. The summary statements on safeguards provisions for Categories III and IV are:

Category III. The material surveillance program for Category III quantities shall assure that when materials are not in locked storage, they are attended, are in authorized locations, and are not accessed by unauthorized persons.

Category IV. The material surveillance program for Category IV quantities shall be site-specific and approved by the Manager, Operations Office."(7)

Thus, Pu-238 is covered by the DOE Graded Safeguards Program, but the safeguards requirements for this isotope are not stringent.

3.0 PROOF-OF-PRINCIPLE DEMONSTRATION

3.1 Demonstration Goals

The advanced concepts demonstration project was set up, using the resources of the ANL Analytical Laboratory, to resolve the proof-of-principle issues associated with this ANL-patented Pu-Be concept for a portable source with separable alpha and neutron-emitter elements. Accordingly, the demonstration goals were straightforward and simple.

They are three in number, as listed in Table 3-1, along with summary statements of the results.

Table 3-1. Proof-of-principle goals and results

1. Demonstrate the deposition of a suitable Pu-238 film in the few-micron thickness range:

A satisfactory 11-mg film of Pu-238 was deposited in the oxide form.

2. Measure the neutron output in the "on" position using a Be plate:

2.5×10^6 n/sec-gm stable output, under vacuum.
Value in good agreement with conceptual design.

3. Measure the neutron output in "off" position:

"Off" emission after two weeks exposure to air was about 5% of "on".
After several weeks air exposure, "off" was stable at about 15%.

3.2 Approach

Surplus Pu-238-powered cardiac pacemakers were used as the source of Pu-238 for the proof-of-principle demonstration. Each pacemaker has oxide particles in the size range of 105 - 250 microns, containing a total of approximately 0.36 g Pu-238, 0.09 g Pu-239, about 0.07 g U-234, a trace of Pu-236, and approximately 0.07 g of oxygen in a triply encapsulated cylinder. (Quantities given have been corrected for decay of Pu-238 into U-234.) The Pu-239, U-234, and O will cause additional absorption of alphas for a given coating mass of Pu-238. Oxygen in the pacemakers is depleted in O-17 and O-18, which greatly reduces the background neutron production by (alpha,n) reactions. However, this depletion cannot be maintained in the PuO₂ planchet deposits, because of necessary intermediate chemical reaction steps that exchange this depleted oxygen for natural oxygen in stock solutions or in the glovebox air.

The ANL Analytical Laboratory used an existing, plutonium air-atmosphere glovebox for preparation of the demonstration planchet deposit, and for radiation measurements of the switch operation. It was recognized that the glovebox did not have a high-purity, noble gas atmosphere. In fact, the glovebox air has no moisture control and is slightly acidic. However, it was desired to perform a convincing and conservative demonstration that a practical SRNS could be made, and a non-purified air glovebox was considered to be suitable for that purpose. Figure 3-1 is a photo of the glovebox. Standard, stainless steel laboratory planchets, 3.25 cm in diam. were used. A photo of the planchet actually used for the demonstration Pu-238 deposition is shown in Fig. 3-2.

The plan of approach was as follows: First, perform a surrogate trial deposition using uranium and a stainless steel planchet. Then, after environmental, health and safety reviews, transfer one triply encapsulated pacemaker source capsule containing about 0.4 grams of Pu-238 in the oxide form (previously radiographed to confirm the physical configuration of the encapsulation), into the glovebox, open the encapsulation, and remove the Pu-238. Next, perform a plutonium trial deposition using plutonium containing no significant fraction of Pu-238. Then, perform the proof-of-principle demonstration deposition of a Pu-238 deposit with thickness of the order of a few microns. The final step is the performance of neutron radiation measurements of background, "on" output, and "off" output, over a period of several weeks; as well as measurements of alpha spectra from the emitter film.

3.3 Experimental Arrangement

A neutron test chamber was designed and constructed to hold the planchet in a horizontal position, deposit side up, for neutron measurements. The chamber contains a Be plate located on a horizontal sliding fixture that can be moved by a pushrod between the "on" position (Be plate over to the planchet) and the "off" position (Be plate not adjacent to the planchet). With the Be plate in the "on" position, the gap between the Pu-238 film and the Be surface was 1.6 mm. The chamber is equipped with a pumpout connection, so that the system can be evacuated to obtain neutron output data without air between the alpha emitter and the light element. Figure 3-3 is a view of the SRNS neutron test chamber.

A 20-cm diameter Bonner sphere Health Physics neutron detector was used to obtain neutron output data. It is not sensitive to gamma-rays and thus registers no response to the 0.775-MeV Pu-238 gammas or the 4.43-MeV gammas from the Be (alpha,n) reaction.

Shown in Fig. 3-4 is a drawing (to scale) of an alpha test chamber constructed to measure the alpha spectrum of the Pu-coated planchet in a vacuum. The chamber is cylindrical with an outer diameter of 10.6 cm. A surface-barrier solid-state detector of 50-mm² detection area views the planchet through a small pinhole and is baffled to prevent any scattered alphas from reaching the detector. This arrangement prevents detector radiation damage and count rate saturation that could be caused by the relatively intense alpha flux from the complete film. In operation, the device sits vertically upright on its base. These measurements were performed with the container evacuated (see shut-off valve). Not shown in the drawing is the mechanism provided to move the pinhole to permit the detector to view a range of positions on the film.

3.4 Experimental Results

3.4.1 Deposit Tests

Electroplating was the original concept selected for this Advanced Concepts study. The concept of electroplating was tested satisfactorily using U-238. This process deposited the uranium as UO₂, not metallic uranium. After the coating was flame-heated, it formed a stable adherent film on the planchet. The next step consisted of performing a plutonium

Fig. 3-1. View of air-atmosphere glovebox.

Fig. 3-2. Planchet to be coated with Pu-238 film.

Fig. 3-3. SRNS neutron test chamber.

Fig. 3-4. SRNS alpha test chamber.

trial electrodeposition in the glovebox using Pu-239. A 25-mg aliquot was taken of NBS SRM 949e (Pu-239) solution, and evaporated to dryness. The residue was dissolved and added to the electroplating cell. After completion of the timed electroplating, the cathode electrode was removed and the planchet examined visually. Material had collected on the planchet, but in the form of poorly adhering, large grains. Because this process did not result in a satisfactory product, a new demonstration procedure was selected, tested successfully using Pu-239, and used successfully for the actual Pu-238 demonstration deposition, as described below.

3.4.2 Demonstration Film Deposition

The alternate method selected was precipitation of the plutonium using oxalic acid ($\text{H}_2\text{C}_2\text{O}_4$). This method was first tested by depositing a Pu-239 film. Again, Pu-239 solution from the NBS SRM 949e material was evaporated to dryness, and dissolved in a 0.05 molar solution of nitric acid. The same apparatus used in the electroplating trial was used for this precipitation. The volume of the plutonium solution was kept low (less than 5 ml) to minimize precipitation of the $\text{Pu}(\text{C}_2\text{O}_4)_2$ on the walls of the vessel. Then, saturated oxalic acid was added to precipitate the plutonium as $\text{Pu}(\text{C}_2\text{O}_4)_2$. After precipitation, the liquid was removed and the planchet with product was air dried. The planchet was then placed in a quartz crucible with cover and placed in a Varian-controlled air atmosphere furnace. The furnace was turned on and heated to about 700C. The rate of ascent was slow, less than 25C/min. After reaching 700C, the furnace was kept on for 3-4 hours and then turned off. This raised the planchet temperature high enough to ensure that the oxalate was converted to the oxide, but no analysis was performed of the film. The planchet was removed from the furnace after cooling, and about four drops of 0.2% CollodionTM in ethyl acetate were added to the film on the planchet, and it was air dried to "fix" the deposit. A visual examination indicated that this deposit was satisfactorily adherent, but the coating was visibly uneven. Then, this process was repeated successfully with Pu-238 from the cardiac pacemaker stock to provide the demonstration Pu-238 film. The net amount of Pu-238 deposited was 11 mg. The total amount of metal was 17.6 mg for material with the composition given in Section 3.2. The surface area of the deposit was about 5.72 cm². This corresponds to a PuO_2 film with an average thickness of about 3 microns.

The demonstration film was deposited on February 1, 1995. Following completion of the baking and CollodionTM coating steps, the planchet was given a visual inspection, and was placed in the neutron test chamber. An initial survey of "on" and "off" neutron emission was performed to confirm a successful plating operation. Then, the planchet was left exposed to the glovebox air atmosphere, except for a few periods of time when the chamber was evacuated using a dedicated mechanical vacuum pump (1-2 hours at a time).

3.4.3 Neutron Emission Measurements

After the film had been exposed for two weeks to the glovebox air atmosphere, neutron emission measurements were performed February 15-16 with air in the neutron test chamber, using an uncalibrated 20-cm diameter Bonner sphere located with its center 20 cm from the center of the planchet, and 50-minute counting periods. The sphere was placed in contact with a circular glove port plastic glove flange, in order to obtain a fixed,

reproducible counting geometry. The "off" emission was $4.3 \pm 2.8 \%$ of the "on" emission. (The uncertainty given is the experimental value of the variance for the counting statistics.)

Additional measurements were taken after further exposure to the glovebox untreated-air atmosphere, between March 22 and March 28, using the same counting geometry and 50-minute counting periods. For these measurements, the sphere was recalibrated using a bare Cf-252 fission source with an emission rate of 7.076×10^6 neutrons/sec. located 50 cm from the center of the sphere. The calibration factor thus obtained was 11.37 counts/minute for a flux of one neutron/cm²-sec.(8)

Results from the measurements taken March 22-28, using the recalibrated Bonner sphere were as follows (uncertainties given are the experimental values of the variances for the counting statistics):

Air in gap:	2.04×10^6	neutrons/sec-gram Pu-238, $\pm 1.3 \%$.
Vacuum in gap:	2.46×10^6	neutrons/sec-gram Pu-238, $\pm 5 \%$.
"off" rate (vacuum):	$13 \pm 1.4 \%$	of the "on" rate.

Visual examination of the inside of the SRNS Test Chamber after completion of these counting measurements showed that a very thin layer had appeared on the back of the beryllium plate. There was no visible contamination on the surface of the Pu-238 film on the planchet, or the surface of the Be plate exposed to the alphas.

Additional neutron emission measurements were made with the Bonner sphere a month later, April 26 and 28, as a check on the effects of continued air exposure. The April data were the same, within counting statistics, as the March results (again, uncertainties given are the experimental values of the variances for the counting statistics):

Air in gap:	2.05×10^6	neutrons/sec-gram Pu-238, $\pm 2.6 \%$.
Vacuum in gap:	2.50×10^6	neutrons/sec-gram Pu-238, $\pm 2.3 \%$.
"off" rate (vacuum):	$14.6 \pm 2.3 \%$	of the "on" rate.

3.4.4 Alpha Spectrum Measurement

Following measurements of the neutron output, a measurement was made of the alpha energy spectrum from the demonstration film, using the alpha test chamber. The detector is an ORTEC solid-state, ion-implanted silicon detector with active area of 50 mm² and a guaranteed maximum resolution of 11 keV full-width-at-half-maximum for 5.486 MeV Am-241 alphas. Figure 3-5 shows the spectrum measured, under vacuum, with the center of the detector positioned approximately over the center of the film deposit. The energy peak is broad and poorly defined, perhaps due to a relatively large microscopic grain size

Fig. 3-5. Energy spectrum of alphas emitted from demonstration Pu-238 film, measured under vacuum.

and/or the visibly non-uniform deposit. However, the spectrum indicates sufficient transmission of high-energy alphas to provide an effective radioactive isotopic neutron source.

3.4.5 Attempt at a Simple Deposition Method to Produce a More Uniform Deposit

Although the $\text{Pu}(\text{C}_2\text{O}_4)_2$ approach was successful, as described above, the film had an uneven appearance. Accordingly, an attempt was made at preparing a film deposit using another, simpler, deposition technique that might produce a more uniform deposit, and was expected to have a significantly smaller grain size. A solution of plutonium in dilute nitric acid was prepared by the same technique described in Section 3.4.1. Drops of this solution were simply added, one drop at a time, to the top of a planchet on a hot plate set at a low temperature, and allowed to evaporate to a dry deposit. The drops were distributed around the planchet to produce as even a deposit as possible. After evaporation of this deposit, the film had a uniform appearance. The planchet then was placed in the quartz crucible with cover, inserted in the Varian-controlled furnace, and treated at 700C, as in the oxalate method. After furnace treatment, no CollodionTM was applied. After the furnace treatment step, the film appearance was significantly finer grained, but more uneven on a microscopic scale, and the deposit was not adherent (see below). Study of this deposition approach was concluded after neutron emission measurements in both the "off" and "on" configurations and an alpha spectrum measurement.

This deposition was performed on July 5, 1995. Neutron emission measurements taken July 17-20, 1995 gave an "off" emission in air that was $14 \pm 3.4\%$ of the "on" emission. However, the emission measured under vacuum on July 20 was less than the air value (measured on July 19). This result was interpreted as indicating that material was lost from the deposit during the chamber evacuation. This interpretation was confirmed by subsequent smear measurements that showed alpha-emitting contamination distributed inside the neutron test chamber, and by subsequent air-atmosphere neutron emission measurements that showed an "on" neutron emission about 28% lower than that measured before the evacuation.

However, the alpha emission spectrum from this deposit, shown in Fig. 3-6, exhibited a sharper energy peak than that from the demonstration film, showing substantially less energy degradation. This result is attributed to the smaller grain size, as compared with the demonstration film. Overall, though, this film did not result in improved neutron output or a lower "off" emission. We suggest this is due to the more uneven distribution on the macroscopic scale.

4.0 DISCUSSION

This demonstration successfully resolved the three proof-of-principle questions it was designed to address. Demonstration goals and results have been presented in Table 3-1. Neutron output is in agreement with an estimate made during development of the concept. Neutron output in the "off" position is low enough for practical use of this concept, although continued exposure to air of the order of days is to be avoided. We cannot, of course, rule

Fig. 3-6. Energy spectrum of alphas emitted from fine-grained Pu-238 film, measured under vacuum.

out chemical changes occurring in the film that were not related to the glovebox atmosphere. The deposition process employed in this demonstration appears to be a satisfactory starting point for producing a practical SRNS. (An attempt to produce a film with more uniform deposit using a simpler technique was not successful.) The data suggest that a practical SRNS should be designed so that the plates are contained in an evacuated capsule, or a capsule filled with an inert atmosphere. Although coated plates prepared by this technique might be stored and used under a vacuum inside the actual source capsule, these results indicate that the plates may be exposed to air for short periods of time. There appears to be potential for reduction of the "off"neutron emission to a few percent of the "on"emission, by special procedures to minimize film contamination. In principle, the "off"neutron emission from a Pu-238 oxide SRNS film could be reduced in this fashion to less than one percent of the "on"emission, but it is not clear that this result could be attained under practical conditions.

The "off"neutron emission from a PuO_2 film coating could be substantially reduced if a process were used that deposited PuO_2 containing oxygen depleted in O-17 and O-18, but development of such a process was considered to be significantly beyond the scope of this project.

Because the Pu-238 neutron emission from spontaneous fission is only about 0.1% of the demonstration film "on"neutron emission, there appears to be potential for reducing the "off"neutron emission fraction to about 0.1% of the "on" emission, by using a metallic emitter film, if there were an application with such a requirement. A metallic Pu-238 film with suitable thickness could be deposited by vacuum-sputtering, followed by storage and assembly operations conducted under vacuum that assured the purity of the metallic film was maintained without oxidation. Such a process would require development effort significantly beyond that employed in this demonstration.

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