MASTER

NEUTRON INTERROGATION OF
LOW-SPECIFIC-ACTIVITY WASTES
WITH A REACTOR

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ABSTRACT

While there have been very concerted efforts to develop passive and active systems for the assay of drums of waste, scrap and/or finished materials, little attention has been given to systems able to assay drums containing fissile materials at low-specific-activity (LSA) levels. For a typical Rocky Flats drum, the Department of Transportation definition of LSA wastes would correspond to ~ 10 mg of plutonium in a drum. These small amounts of material are not particularly important from the viewpoint of accountability. However, LSA drums should be assayed from a safeguards viewpoint to insure that material has not been accidentally or covertly placed in LSA drums. They should be assayed from an environmental viewpoint to insure that there will not be an accidental release of material to the environment during waste disposal. Calculations will be presented to demonstrate the feasibility of detecting 1 to 10 mg quantities using a multipurpose reactor. An experimental verification of this feasibility has begun. Results available at the time of this meeting are presented.

INTRODUCTION

Safeguards and accountability, which are the primary concerns of this meeting, will not be neglected in this paper. However, we wish to put our emphasis on the environmental problems which all of us in the nuclear industry must face. The Rocky Flats Plant has unique problems, and we are studying an unusual solution to one of them. We are addressing ourselves to the problem of low-specific-activity (LSA) wastes.

Considerable effort has been, and will be, expended on solutions to the problems of safeguards, accountability, and disposal of high-activity-level waste. Underground storage or burial in remote locations, or at sea, may suffice for the disposal of high-level wastes. However, public acceptance is not complete for present and proposed storage locations. These problems are under intensive research, and solutions must undoubtedly be found.

We believe that the next problem to which the nuclear industry will be subjected is that of its low-specific-activity (LSA) waste. It is important to be able to state that LSA waste is indeed LSA waste. Most LSA waste at the Rocky Flats Plant contains no radioactivity because it was generated in offices, cafeterias and other places where no radioactivity should exist. Other LSA waste drums are generated inside production areas, but are defined as LSA waste because of the function being

served in that part of the production area. Some drums may actually contain LSA levels of plutonium. Consider that accidents in use of disposal drums can, and probably will, happen eventually. Perhaps a drum might be mistakenly labeled as "LSA Waste".

How does one prove waste to be "LSA"? The answer to this question will vary with each plant processing fissile and/or fertile materials. In any case, we believe that governmental agencies, and the public, will eventually want such proof. There is also economic justification for proof of LSA levels because it is easier and cheaper to dispose of LSA wastes. With environmental pressures mounting, and with economic justification for inexpensive transportation of LSA wastes, how can a facility with the mission of the Rocky Flats Plant meet its LSA-waste-assay problem?

We shall briefly consider several facets of this problem at Rocky Flats to provide some understanding of one proposed solution. For purposes of this discussion, we have chosen to use the Department of Transportation definition of LSA wastes which is 0.1 μCi per gram of plutonium-contaminated waste. In typical Rocky Flats 55-gallon drums, this activity level corresponds to one to ten milligrams of our plutonium.

The Rocky Flats Plant is unique in the sense that it processes plutonium with an approximately constant relative isotopic composition. Since our plutonium contains appreciable amounts

of plutonium isotopes other than ^{239}Pu , there can be complications in its assay. For example, there might be appreciable amounts of the ^{241}Am product of decay of ^{241}Pu which is present in our plutonium. The spontaneous fission neutrons from ^{240}Pu , which is also present in our plutonium, provide a sometimes useful signature in addition to the gamma-rays emitted by the major isotope ^{239}Pu . The chemical form is important. Plutonium metal, PuO_2 , PuOF_2 , PuF_4 and PuF_3 exist in the plant. The plutonium is sometimes intimately mixed with low-atomic-number metal salts. Each chemical form will provide a different neutron output per gram of plutonium because of (α, n) reactions induced in the low-atomic-number elements present with the plutonium.

Speaking for Rocky Flats, one can say that the assay problems are recognized. Our problems are simplified by the approximately constant isotopic composition of the plutonium which is processed in the plant. We know that ISA levels of plutonium in 55-gallon drums can be assayed by passive systems, using NaI(Tl) , or even Ge(Li) detector systems. Long counting times, ranging from 5 to 60 minutes, would be required. We also recognize that passive gamma-ray signatures can be obliterated by intentional or unintentional shielding, e.g. by lead-loaded glovebox gloves or by metals such as iron piping.

The last statement is also true for neutron interrogation. Neutron penetration and/or exit from a drum would be strongly influenced by the amount of low-atomic-weight elements present in a drum. Hydrogenous materials are particularly troublesome

for low-energy neutron interrogation. However, much of the waste from Rocky Flats has low density. Computational estimates convinced us that our low-density LSA wastes, which contain hydrogenous materials such as papers, plastics and cloth, could be interrogated using a relatively "soft" neutron beam. Waste categories with high density and/or atomic number are more amenable to neutron interrogation, provided that the dominant material in the waste or scrap does not have an exceedingly high scattering or absorption cross-section for neutrons. Iron or stainless steel piping, loosely placed in a drum, would satisfy the requirements for neutron interrogation, whereas gamma-ray attenuation makes this type of waste, or scrap, difficult to assay by passive gamma-ray systems.

We have followed closely the experimental and theoretical studies at other laboratories. The groups at these facilities are providing valuable services and solutions to the nuclear industry's assay problems. Because our requirements have a specific nature, we have developed our counting systems in-house. The in-house developed passive gamma-ray counting systems at Rocky Flats suffice for about 50% of our assay requirements. These are drums containing high-activity-levels of plutonium. The other 50% of our assay requirements involve high density matrices (e.g. metal scrap), but particularly LSA waste.

Another aspect of the problem is the number of LSA drums generated, which amount to several thousand per year, as well as several hundred crates per year. An LSA assay system

must have a high throughput capability. We estimate an irradiation-plus-measurement time of ~ 100 - 120 secs can be achieved. Total drum handling time should be less than 5 mins. Passive systems cannot, in general, perform this service on all categories of our waste at LSA levels.

Bringing all these considerations together, we felt that a reactor which was originally proposed for other analytical and non-destructive-testing purposes at Rocky Flats, might serve an additional important function in the assay of LSA wastes and of metal-category wastes.

A reactor is an extremely potent neutron source, and a reactor compares favorably with other methods for the problem under discussion. The main advantages for this problem are:

- 1) high steady-state neutron output at constant power levels for extended periods of time;
- 2) reliability;
- 3) pulsing capability can be provided; and
- 4) for reactors using TRIGA-type fuel and configuration, excellent safety.

Reactors are not trivial devices to operate; however, there were fourteen prospective uses for a reactor before we decided to investigate the feasibility of LSA waste detection.

Practical geometrical and mechanical considerations of installing a drum-sized beam port would probably limit the deliverable steady-state flux to $\sim 2 \times 10^8 \text{ n}/(\text{cm}^2 \text{ sec})$ thermal neutrons and a similar number of fast neutrons, depending upon the beam tailoring. This flux could be delivered fairly uniformly to an entire drum for a total of $\geq 10^{13} \text{ n/sec}$. With this flux, one could detect $\sim 0.25 \text{ mg}$ of plutonium in a drum. If pulsed, $\sim 0.01 \text{ mg}$ is the practical theoretical limit because of the increase in sensitivity obtained by pulsing⁽¹⁾. This may be contrasted with a reported⁽²⁾ sensitivity of 50 mg of ^{235}U for a pulsed-neutron-generator system with a 100 sec scan time. Much longer scans would be required to achieve a 1 mg sensitivity for ^{235}U or ^{239}Pu . Isotopic sources, e.g. ^{252}Cf , could also be used. These sources suffer a disadvantage of decay and therefore decreasing source strength, so that ~ 0.01 to 0.03 g of ^{252}Cf would be required to have a long, useful life of ~ 5 to 8 years. This source would not fulfill other needs which the reactor can provide. Linear accelerators also produce copious amounts of neutrons, but are expensive and do not provide the versatility of simultaneous irradiations and other beam-port experiments which a reactor can provide. In short, a reactor is more versatile and can assume more simultaneous functions than the other sources mentioned.

EXPERIMENTAL PROCEDURES, CALCULATIONS AND RESULTS

The method under consideration is active interrogation with a drum-sized neutron beam from a reactor, followed by delayed neutron counting at a remote location.

Our investigational program, in collaboration with Gulf Radiation Technology, consists of several parts. First, we have initiated a calculational effort for 55-gallon-sized containers. Second, we have investigated the response of a small-scale system with plutonium samples in 3-quart or gallon-sized containers. Third, we have performed flux-mapping experiments with 55-gallon drums, using mocked standards of our waste categories. Fourth, we are in the process of simulating in small-sized containers a flux-mapping experiment which can be extrapolated to the 55-gallon drum size.

Fifth, we have performed calculational estimates of the residual activities in a drum and its contents to verify that irradiation, followed by a suitable "cooling" period, does not significantly increase the activity level of the drum and its contents.

The authors considered several ways of estimating the flux required to do the required task. A straightforward calculation provided an estimate that $1.4 \times 10^7 \text{ n/cm}^2 \text{ sec}$ would suffice to detect 10 mg of plutonium. Approximately $1.4 \times 10^8 \text{ n/cm}^2 \text{-sec}$ should detect 1 mg of plutonium in a 55-gallon drum.

The equation used was:

$$\phi = \frac{(N-B)}{t_b \cdot \sigma \cdot nx \cdot Y \cdot \xi \cdot l \cdot \sum_i f_i (1 - e^{-\lambda_i t_b}) (e^{-\lambda_i t_a} - e^{-\lambda_i t_c})} \quad (1)$$

where:

N = Total neutron count.

B = Neutron background.

t_b = Bombardment time of the drum.

t_a = Time after exposure before counting begins.

t_c = Time after exposure when the counting ends.

σ = Fission cross-section.

nx = Number of atoms of ^{239}Pu in the drum.

Y = Absolute yield of delayed neutrons per fission.

λ_i = Decay constant for each component of the delayed neutron spectrum.

ξ = Detection efficiency.

l = Fractional leakage of delayed neutrons from the drum.

Values assumed for the calculation are $(N-B) = 500$, $t_b = 25$ sec., $t_a = 5$ sec., $t_c = 55$ sec., $\sigma = 740$ barns, $nx = 2.52 \times 10^{18}$ (the number of atoms in 10 mg of ^{239}Pu), $Y = 6.1 \times 10^{-3}$, $\xi = 0.1$, $l = 0.5$.

The values of λ_i may be calculated from the relation $\lambda = \ln 2/T_{1/2}$.

The reported neutron groups (3) present in the delayed neutron spectrum are given in Table I, along with the fractional abundance f_i , and the products $f_i (1 - e^{-\lambda_i t_b}) (e^{-\lambda_i t_a} - e^{-\lambda_i t_c})$. It is clear that one should choose the irradiation and counting times to utilize the most abundant groups with half-lives from 2.1 to 23 secs.

TABLE I

i	$T_{1/2}$	Fractional Abundance, f_i	The Product
			$f_i(1-e^{-\lambda_1 t_b})(e^{-\lambda_1 t_a}-e^{-\lambda_1 t_c})$
1	54.28	0.034	0.0041
2	23.04	0.299	0.1058
3	5.6	0.212	0.1086
4	2.13	0.326	0.0632
5	0.62	0.085	0.0003
6	0.257	0.044	0.0000

In addition, one-dimensional, neutron-transport calculations, using the code 1DF, were performed for us at Gulf Radiation Technology for two categories of our waste, dry combustibles and wet combustibles. Dry combustibles were chosen because this category will simulate the largest number of LSA waste drums containing paper, rags, rubber gloves, etc. The second category was chosen to represent a "poor" case for low-energy-neutron interrogation, because the matrix is 52 per cent water which will severely attenuate and scatter low energy neutrons. The results are shown in Table II. These results are for a homogeneous source of 10 mg of plutonium. If these normalized delayed neutron leakages are multiplied by the area of a drum, i.e. $\sim 4800 \text{ cm}^2$, and it is assumed that a 10 per cent efficient counting geometry is available and that 30 per cent of the delayed neutrons will be emitted during the counting period, then there should be a count of ~ 700 neutrons. This compares favorably with the 500 or so counts which we assumed would be needed in our simplified calculation, using Eq. 1. The essential agreement of the two results is encouraging.

TABLE II

Results of 1DF Barrel Calculations (10 mg Pu²³⁹)

CASE	Prompt Fissions*	Delayed Leakage*
WET	2.22×10^{-5}	5.62×10^{-8}
DRY	3.39×10^{-5}	1.33×10^{-7}

* per incident neutron per unit area integrated over the source energy spectrum

Experimental verification work has begun at Gulf Radiation Technology in San Diego, using the Mark I TRIGA reactor. The earliest results of this effort are highly promising.

The initial experiment is illustrated in Figure 3. The sample consisted of 9.8 mg of 95% ^{239}Pu as PuO_2 distributed in diatomaceous earth. The sample was placed in a 2" thick paraffin-walled neutron-reflecting cavity with inner dimensions of 7 in. x 7 in. x 12 in. This was, in turn, shielded by $1/2$ in. of lead, 0.020 in. of cadmium and $1/2$ in. of boron-loaded plastic. The entrant beam had a flux of $1.0 \times 10^7 \text{ n}/(\text{cm}^2\text{-sec.})$. After a 60-second irradiation, the reactor was scrammed and the sample was hand-carried to the counting geometry by running with it. This transfer normally required ~ 7 to 9 secs. before the top cover was closed and delayed-neutron counting began. Counting was continued for 60 secs. The counting geometry consisted of $6\text{-}3/4$ in. diameter by a 16 in. deep cavity surrounded by eight ^3He -detectors imbedded in the polyethylene shield. This geometry was further shielded from the delayed neutrons emanating from the reactor by 12 inches of boric-acid-filled boxes, on five sides. It was shielded on the top by $3/8$ in. of boron-loaded plastic and 0.020 in. of Cd. The counting geometry was physically located ~ 10 feet from the reactor in the same room.

The results of this experiment gave a signal to background ratio better than 10:1, i.e. 500 counts: 30 counts. Most important is the extrapolation which can be made. A similarly constructed geometry, far removed and further shielded by concrete walls, should have been able to detect ~ 0.1 mg of plutonium, since

the background of the counting system was demonstrated to have a background ~ 20 times lower than the reactor-scrammed delayed-neutron background.

A small 10 mg sample was moved about in an empty can inside the reflector, with a variation in counts of $\sim 20\%$. Similar experiments in a paraffin-filled container showed that the paraffin would increase the detectable limit to ~ 0.25 mg of plutonium, using the parameters of this experiment. There was a considerable variation of a factor of 3, depending on the position of the point source in paraffin.

While these extrapolations are gratifying in that they met, or exceeded, our expectations, we proceeded to investigate the problem of full-sized drums with dry-combustible and wet-combustible categories. Representative standards were constructed at Gulf Radiation Technology according to prescriptions provided by the Rocky Flats Standards Laboratories. These standards contain 27 one-gallon nalgene jars and 3 one-half-gallon spacers. Dry combustibles contain some empty jars to achieve the proper density.

Manganese monitors were inserted in the drums. One hour irradiations were made and the monitors were counted to obtain a "poor" geometry flux map. The geometry is shown in Figure 4, which is a top (or bottom) view of a standard 55-gallon drum over the

8 in. x 10 in. beam tube available at the Gulf Mark I TRIGA. The results of the irradiations are shown in Figure 5 for dry combustibles. Monitors were placed only toward one end and side of the drum, since the drum was symmetrically placed over the beam tube. The dry-combustibles drum results may look slightly unusual, since a jar of matrix nested next to an empty jar at the beam entry area. This resulted in a slight sideward scatter and a slight decrease in the flux directly upward along the center of the drum. Figure 6 shows the wet combustible case in which there was a matrix in each jar. The results are more regular here.

Two important lessons were learned from these experiments. First, the worst case that one is likely to encounter is that the flux at the center is ~ 15 times lower than the entrant flux. This will not occur if the entire drum is irradiated. Conservative scattering of neutrons entering the edges rather than the central portion of the drum will increase the flux in the central portion. If a drum is rotated, particularly within a reflective cavity, all outer portions of a drum will be exposed to the flux. Finally, beam "tailoring" toward a "harder" flux can help mitigate the problem. The next important lesson was that many experiments would be required to entirely map and measure the contributions from each portion of a full-sized drum if one has a relatively small beam port.

For the fourth part of the program, we have begun to mock drums in miniature size, using wood and plastics to achieve the proper atomic densities (principally the hydrogen density) and thus the proper relative scattering lengths. These model drums can be exposed in a uniform flux with the beam part available at the TRIGA reactor. These data are not available for presentation at this time. However, an entrant flux of $\sim 5 \times 10^8$ neutrons/(cm².sec) should still be able to interrogate the central regions of a drum.

Although the fourth part of the program is not completed, we have performed sufficient calculations to satisfy ourselves that we shall not contribute enough additional activity to the waste to be of any significance. Ninety-seven slow-and fast-neutron reactions, i.e. (n, γ), (n,2n), (n,n'), (n,p) and (n, α) reactions on the isotopes of sixteen elements most likely to be found in Rocky Flats wastes were considered. Our conclusion is that 24 hours after a 1-minute irradiation at 10^8 n/(cm².sec.) the drums and ISA contents will have returned to background levels. Indeed, ~ 2 hours after irradiation, they will, in general, meet ISA definitions for all types of radiation.

SUMMARY

The calculational and experimental results to date show every indication that ISA levels of plutonium down to 1 mg in a 55-gallon drum, can be detected with a high degree of certainty.

Our immediate efforts for the future will be directed toward identifying extreme cases in which the plutonium might "hide" from interrogation, what beam-tailoring characteristics would be most desirable, what reflector characteristics are needed, and the best means to transport drums into an acceptable counting geometry.

Lest we be criticized for swatting a fly with a football-field-sized fly swatter, we wish to point out that our yearly drum output could nearly cover a football field.

ACKNOWLEDGEMENTS

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FIGURE CAPTIONS

1. Proposed Flow of Waste Drums with Reactor Verification of Plutonium Content.
2. Conceptual Design of Activation Analysis Facility - Rocky Flats.
3. Illustration of Pu Sensitivity Experiment for Gallon-Sized Containers.
4. Top View of Flux Mapping Irradiation.
5. Relative Flux Map of Dry Combustibles Drum Filled with Mock Dry-Combustibles Category Waste Taken at 8" x 10" Beam Port.
6. Relative Flux Map of Wet Combustibles Drum Filled with Wet-Combustibles Category Waste Taken at 8" x 10" Beam Port.

PROPOSED FLOW OF WASTE DRUMS

ACTIVATION ANALYSIS FACILITY - ROCKY FLATS

ADMINISTRATION & LABORATORY BLD'G

REACTOR BUILDING

CONTROL ROOM

UTILITY AREA

REACTOR ROOM

REACTOR

DRUM COUNTING

FY 1973 ITEM

14,600 SQ. FT.

CONCRETE STRUCTURE

JUSTIFICATION

ACCOUNTABILITY

DETERMINE AMOUNT OF PLUTONIUM
IN WASTE

SAFEGUARDS

POSITIVE CONTROL OF RADIOACTIVE
WASTE

NEUTRON RADIOGRAPHY

NON-DESTRUCTIVE TESTING OF
VARIOUS COMPONENTS

ELEMENTAL ANALYSIS

ANALYSIS OF PLANT EFFLUENTS AND
SURROUNDING ENVIRONMENT

FY 1973 ITEM

14,400 SQ. FT.

CONCRETE STRUCTURE

JUSTIFICATION

ACCOUNTABILITY

DETERMINE AMOUNT OF PLUTONIUM
IN WASTE

SAFEGUARDS

POSITIVE CONTROL OF RADIOACTIVE
WASTE

NEUTRON RADIOGRAPHY

NON-DESTRUCTIVE TESTING OF
VARIOUS COMPONENTS

ELEMENTAL ANALYSIS

ANALYSIS OF PLANT EFFLUENTS AND
SURROUNDING ENVIROMENT

N

Pu SENSITIVITY EXPERIMENT

TOP VIEW OF FLUX
MAPPING IRRADIATION

FLUX MAP OF DRY COMBUSTIBLES DRUM

END

1/4

CENTER

1/4

END

RELATIVE FLUX

BEAM TUBE

PROJECTIONS OF MATRIX-CONTAINING JARS ON END OF DRUM.

FLUX MAP OF WET COMBUSTIBLES DRUM