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STORAGE OF PLUTONIUM

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V. R. Cooper

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STORAGE OF PLUTONIUM

by

V. R. Cooper  
Acting General Manager

September 29, 1959

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HANFORD ATOMIC PRODUCTS OPERATION

CHEMICAL PROCESSING DEPARTMENT . . RICHLAND, WASHINGTON . . TELEPHONE 2-1111

September 29, 1959

U. S. Atomic Energy Commission  
Hanford Operations Office  
Richland, Washington

Attention: Mr. A. T. Gifford, Director  
Process Engineering and Manufacturing Division

Gentlemen:

STORAGE OF PLUTONIUM

Reference: Letter, A. T. Gifford to W. K. MacCready  
"Storage of Plutonium", dated  
September 22, 1959 (OS:EBJ)

In response to your referenced request, we have reviewed the available information bearing on your specific questions about the storage characteristics of plutonium, and the pertinent information is summarized below. For convenience, we have presented the information in the form of direct answers to the questions posed.

1. Stability of plutonium nitrate solutions

Relatively pure acid-plutonium nitrate solutions, such as those produced by an anion exchange purification process, are quite stable and may be stored satisfactorily at ambient temperature for long periods of time. A minimum acidity is required to prevent the polymerization and precipitation of plutonium; for example, a solution of ten grams plutonium per liter is stable at 0.7 molar nitric acid, while a solution of 200 grams plutonium per liter is stable at 2.5 molar nitric acid. The precipitates formed in solutions containing less than the critical concentration of acid are of indefinite composition but are probably hydrous oxides; they are quite refractory, especially if exposed to temperatures higher than ambient, and require extended boiling in nitric acid (and sometimes the addition of fluoride ion) to dissolve.

Some gas is generated during the storage of plutonium nitrate solutions. In addition to the helium from alpha decay, some oxygen may be liberated via the chemical reduction of plutonium

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from the hexa to the tetra-valent state. Some decomposition of water may also occur. Storage in sealed containers may be permitted for a few months provided a relatively large vapor space is included to permit the accumulation of gas at low pressures. A vented vessel should be used if the solution is to be stored for an extended time.

The storage conditions outlined above are applicable for plutonium of any isotopic ratio likely to be encountered in spent reactor fuels.

## 2. Alternative forms for storage

### a. Buttons

Plutonium metal buttons can be stored in metal cans for extended periods of time, provided certain conditions are met. Sufficient free space or venting must be provided to allow for storage the gas resulting from radioactive decay. The atmosphere should have a dewpoint of  $-20^{\circ}$  C or lower, and an oxygen content of perhaps 2-5% to avoid oxidation of the plutonium. The can must be so designed as to dissipate the heat of radioactive decay at a temperature compatible with the other criteria. These conditions can be met in either a controlled atmosphere hood or in sealed metal containers. If the buttons are sealed in cans, periodic inspection should be made to assure the continuing integrity of the container.

### b. Oxide

Plutonium oxide is chemically quite stable. No special precautions need be taken with respect to atmosphere, and the generated gasses may be readily vented as produced. A slab type container would be desirable to facilitate the dissipation of the heat of radioactive decay. The handling of dry powders is very undesirable because of problems resulting from air-borne contamination.

### c. Fluorides

In general, the comments concerning oxides are equally applicable to fluorides. An additional complication stems from the appreciably increased neutron flux resulting from the  $\alpha$ -n reaction with fluorine.

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## d. Solutions

The storage of nitrate solutions is feasible so long as certain precautions are observed. The storage vessel would need to be geometrically safe (about a 1.5" slab or a 5" diameter cylinder) or each batch would be limited to about 250 grams plutonium. These values presume a true solution of no more than 500 grams plutonium per liter. Each proposed storage plan must be specifically evaluated to assure nuclear safety. The acidity must be controlled to prevent precipitation and the containers should be vented in a way that prevents undue loss of water or acid through evaporation. The dissipation of heat through the vessel walls could be effected without unduly complicating the vessel design. At ambient temperatures, the corrosion of stainless steel would be negligible.

## e. Slurries

The characteristics and limitations of slurries in storage are quite similar to those of solutions, except that the acidity need not be controlled so closely. If temperatures higher than ambient are encountered, the precipitates are likely to be converted to a refractory form.

## 3. Ease of subsequent processing

Plutonium nitrate solution can be readily processed for the removal of Am-241 or U-237 or for conversion to other chemical forms by existing technology. Other forms of plutonium must currently be converted to a plutonium solution prior to the removal of decay daughters. Any of the chemical forms can be processed to the metal by existing technology.

It appears probable that the technology of the future will make greater use of pyro-processing techniques. In this event, the metal might be the form most compatible with the then current technology.

## 4. Reproducibility of assay

## a. Nitrate solution

Representative samples of plutonium nitrate could be readily obtained from storage vessels and assayed by existing technology. The mass of the stored material could be readily verified, permitting periodic verification of inventory.

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## b. Oxide

Samples of plutonium oxide could be readily obtained and the mass of the stored material readily determined. The chemical composition and uniformity of the stored material would be known only if the mass were heated in the presence of air to about 1000° C prior to storage. This technique would permit reproducible assays, but would increase the refractory nature of the material thereby complicating subsequent processing.

## c. Fluoride

Verification of inventories would be inexact and unpredictable, because of the indefinite composition of the fluoride salts.

## d. Slurries

The verification of an inventory of stored slurries would be virtually impossible without processing the material to a known compound in either a solution or dry state.

## e. Metal

The mass and assay of a plutonium metal button could be verified readily. Buttons stored in a controlled atmosphere hood could be inventoried readily. Buttons stored in sealed metal containers could be verified by total mass (button plus container). If absolute verification were required, the container could be opened in a controlled atmosphere hood, the button weighed and sampled, and the button resealed in another container.

## 5. Incremental costs

We are unable to estimate the incremental costs of performing the indicated chemical operations, since these costs are unique to each specific operating situation. The directly variable costs, which include only the cost of essential materials and utilities, can be estimated and are presented below.

Converting nitrate to buttons	\$35.00 per kilo. plutonium
Converting buttons to nitrate	\$ 0.51 " " "
Converting nitrate to oxide	\$ 1.84 " " "

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Converting oxide to nitrate \* \$ 4.25 per kilo. plutonium

\* This nitrate solution would contain relatively high concentrations of other ions and would have to be chemically purified prior to recovery of the plutonium in usable form.

The significant cost of conducting any of the above operations is associated with operating and service personnel and with the provision and maintenance of the required equipment. The operating and maintenance costs are primarily functions of time and could be expected to increase stepwise with increasing quantities of plutonium processed.

Estimates of these incremental costs must be based on rather specific assumptions as to the magnitude, timing and character of the incremental load, and the other work in progress at the time.

On the whole, we would prefer to store plutonium as buttons, sealed in aluminum cans. In this form, the material is stable, readily inventoried, and readily processed into other desired forms. Should you desire more quantitative information as to costs or specific storage conditions, we would be pleased to develop additional information specific to the proposed operating conditions.

Very truly yours,

ORIGINAL SIGNED BY  
A. B. COOPERActing General Manager  
Chemical Processing Department

VR Cooper:RET:wb

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