

W. B. Creamer

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MOUND LABORATORY - MAMMANTO
Central File No. 63-8-215

August 13, 1963

Mr. W. B. Creamer, Area Manager
U.S. Atomic Energy Commission
P.O. Box 66
Miamisburg, Ohio

Dear Mr. Creamer:

Shipment of Pu-238 from Savannah River
Plant to Mound Laboratory.

To date Mound Laboratory has received 3,504.51 liters of plutonium nitrate solution containing 12,303.1 grams of plutonium-238. This material received in a total of 52 different shipments has been transported and unloaded without incident. At the beginning of the program which required the transportation of plutonium-238, various systems of containment and forms of material were carefully evaluated. At that time it was determined on the basis of safety and total cost that the best way to prepare the material for shipment to transport it over the road, and to receive the material was as a nitrate solution. The solution has the following advantages:

1. Spillage of the solution does not present a serious health physics problem. The plutonium nitrate in solution is not airborne until after it has been completely dried out. Since this solution is acidified with nitric acid, the liquor on drying passes through a series of hydrated complex nitrate salts. These salts are deliquescent. As a result, the material arrives to a dusting condition very slowly. The experience at

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Mound Laboratory to date has shown that spillage of a nitrate solution does not present a serious health physics problem. Decontamination with water and complexing agents is readily accomplished before the material becomes airborne.

Mound Laboratory has had no health physics problems involving personnel arising from spillage of nitrate solutions. Personnel have been contaminated externally and internally with nitrate solutions. External contamination not readily removed by scrubbing with decontaminating agents safe to use on human skin has been isolated until new skin formed through natural processes and the contamination was removed through normal skin sloughing. Internal contamination acquired by body uptake of the material has been excreted rapidly without showing the long term retention characteristic of the insoluble material.

2. The plutonium nitrate solution is readily convertible to any desired form such as oxide, metal or salt. The chemical processing of the nitrate solution to any desired chemical form is relatively easy. Since the material is in solution, conversion to a solid through evaporation and low temperature ignition, precipitation of the plutonium as the oxalate, the peroxide or the potassium sulfate double salt, or reduction to the metal are straight forward chemical processing operations with known reaction losses. The resultant material in waste streams is in known concentration and readily recovered by solution chemistry.
3. Storage of the plutonium nitrate solution is safe. Stainless steel containers are not destructively corroded by the nitrate solution. Normal nitrate passivation of stainless steel by the concentration of nitrate iron in these solutions is sufficient to withstand the character of the plutonium-238 nitrate

system. Container problems have occurred only in those situations where the solutions contain halogen ions which had been added to accomplish the solution of oxides of plutonium. The gases resulting from the radiation decomposition of water are readily vented or may be recombined using catalytic recombiners.

Current planning is directed toward the preparation of plutonium oxide at the Savannah River Plant and its distribution in unloadable containers. The plutonium oxide has the following serious disadvantages which should be carefully evaluated before monies are spent to provide oxide preparation facilities at Savannah River Plant, adequately safe containers to ship the material and oxide handling facilities, both physical and chemical, at Mound Laboratory.

1. Plutonium oxide is a serious health physics hazard. Since a considerable amount of the solid plutonium oxide occurs as fines of a submicron size, it is readily airborne. Any release will produce a serious and costly contamination problem. A serious transportation accident may result in the contamination of many square miles of countryside. Any accident during loading or unloading of the container may result in the contamination of an entire facility.

The serious hazard to all operating personnel also must be considered. Plutonium oxide is insoluble in normal body fluids. All medical history to date has been negative in attempts to increase the solubility of this material after biological ingestion. All known serious body burden cases have involved the plutonium in a biologically insoluble form as a lung burden. The material as an insoluble oxide residing in the lungs thus becomes a continuing supply to the body as extremely slow solution is effected. The studies at the Hanford Laboratories confirm these statements. Deaths to laboratory dogs were

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caused by respiratory failures resulting from fibrosis of the lungs due to continued alpha exposure.

2. Plutonium oxide is difficult to dissolve both from chemical considerations and health physics aspects. The difficulty of dissolving plutonium oxide is directly related to its previous thermal history. Plutonium oxide precipitated from an oxalate system must be calcined to insure the completion of the decomposition of the oxalate before packaging to prevent pressurization of the container during storage and shipment. The dissolving of the calcined oxide required a boiling digestion in concentrated nitric acid containing a small amount of fluoride. The fumes from such a reaction cause rapid deterioration of all duct work and mechanical equipment associated with exhaust system. Elaborate scrubbers must be installed along with condenser systems to reduce this deterioration. The necessary fluoride concentration will increase the neutron flux during this operation. It is expected that the necessary personnel shielding will require the installation of remote handling equipment.
3. Solution of calcined plutonium oxide in nitric acid-fluoride media is extremely slow and often does not go to completion. The remanent oxide must be separated from the solution and solubilized by high temperature salt fusions or possibly by a high temperature chlorination. Either one of these methods is extremely slow and costly. Batch sizes must be small. Purification operations on the products of either operation results in large quantities of waste solutions which require additional processing.

It is requested that the proposed transfers of plutonium-238 as its oxide be carefully reviewed in light of the above statements. The shipment of the oxide will require the receiver to convert the material back to the nitrate - the same

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chemical form as available at the Savannah River Plant prior to the oxalate precipitation and calcination. It is our judgement that the proposal to ship the plutonium-238 as an oxide rather than as a nitrate solution will constitute a serious health and safety hazard as well as being not economically feasible.

If you have any further questions on this matter, please do not hesitate to contact me.

Very truly yours,

Original signed by D. L. Scott

David L. Scott
Vice-President,
Plant Manager

LVJ/DLS:cam

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