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PLUTONIUM FOR RECOVERY FROM SCRAP
AT ATLANTIC RICHFIELD HANFORD COMPANY

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PLUTONIUM FOR RECOVERY FROM SCRAP
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ABSTRACT

Operating procedures used to receive and convert plutonium-bearing scrap to high-purity plutonium nitrate product are discussed.

Operating procedures used to measure or verify, repackage, and store are also discussed.

Shipper obligations or responsibilities and receiver handling methods are described.

Nuclear Materials Management procedures pertinent to accounting, measurements, and inventory control are outlined.

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INTRODUCTION

Since September 1966, Atlantic Richfield Hanford Company, or "ARHCO," has had the responsibility for operating the chemical processing portion of the Atomic Energy Commission's work conducted at Hanford. In prior years, this same work was performed by other contractors. For those who are not acquainted, our operations are located in the southeastern part of the State of Washington and are an integral part of the overall Hanford complex.

The particular plant under discussion today is known as the Plutonium Reclamation Operation. Its function is to recover and refine plutonium-bearing scrap. Our objective is to briefly describe the production activities and to outline the techniques that are employed to control the various inventory accounts. The Plutonium Reclamation Operation is subdivided into several material-control centers. Chart I depicts the respective centers with product flow lines showing the progress of scrap from the point of receiving to a final concentrated and purified product. Subsequent charts deal with individual control centers as indicated below.

- II. Feed Stock
- III. Incinerator and Leaching
- IV. Solids Dissolution
- V. Solvent Extraction
- VI. Unique Processes

FEED STOCK

Chart II relates specifically to the Feedstock Control Center. The plutonium-bearing scrap items stored in these locations are packaged in metal

containers, and placed in approved arrays. These inventory storage locations are widespread. For instance, the inactive feedstock is stored in seven Army quonset huts located eight miles from the processing plants. The active feedstock is located near the processing plants, in another Army quonset hut. Within the plant proper, the feed is stored in specified vaults, rooms, docks, and corridor locations.

To give an idea of order of magnitude, consider that the feed inventory of plutonium-bearing scrap awaiting recovery comprises many thousands of packages. These packages have plutonium values that range from as little as 10 grams to 2 kilograms each. In contrast, the gross weight of all this scrap could be measured in tons.

For convenience in identification grouping and process matching, the feedstock inventory is divided into 39 categories of plutonium scrap, located in up to 59 different specific inventory locations.

The accounting for this complex arrangement is performed manually, with assistance from an electronic data-processing system that has functioned for a number of years. The system is programmed to identify the inactive items in the outlying quonset huts by specific hut location, lot, item number, and by associated grams of plutonium. All items in the various active inventory locations are also controlled by specific location, item number, and associated grams of plutonium.

Receipt, removal, and adjustment data involving activity in all feedstock locations are keypunched and processed through an IBM 7090, on a monthly basis. This monthly updated EDP inventory output gives the revised status of each item in the feedstock inventory by its respective item number, category, physical location, and associated grams of plutonium--summarizing the total feed inventory.

A physical inventory is made monthly, to comply with AEC requirements. This monthly physical inventory is made only in the active inventory locations and is recorded on forms that are adapted to keypunching. The monthly inventory in the outlying inactive storage areas is by book. However, an annual inventory is taken of inactive items, using statistical sampling techniques.

Movements into and out of these locations are recorded as they occur, and are blended into the total EDP system. The system is programmed to provide complete reconciliation. In consequence, the output occasionally reveals items omitted or items in excess for reconciliation. Reconciliation effort is immediately expended.

It is of particular importance to note that the receipt measurement of plutonium, and the description of the material contained in a scrap package, is the responsibility of the originating facility. The verification measurement capability of the receiver is limited to item count, gross-weight check, and the verification of the presence of plutonium by gamma scan or neutron count.

Instrumentation has not yet been developed and tested which can be used by the receiver to measure plutonium to the desired degree of accuracy for all of the varieties, combinations, and conditions of plutonium-contaminated scrap materials.

The receiving plutonium measurement for a specific feed item remains identified with the item throughout its stay in the feed inventory. Removals from the Feedstock Material Control Center to the Processing Material Control Centers are transferred, using the plutonium values assigned by the originator.

INCINERATION AND LEACHING

Chart III references incineration and leaching, or the next Material Control Center. This operation is the first of the processing activities to be described. Typical feed to this control center is in the form of plutonium-contaminated dry-box gloves, rags, filters, kimwipes, plastics, tools, piping, and insulation that are packaged in large cardboard waste cartons (size 18 1/2" x 18 1/2" x 24 1/2").

Waste carton contents are transferred to the hood, based on originally assigned plutonium value. A neutron count is made to confirm these prior recorded measurement values. Items contained in the carton are hand-sorted in a dry glove box to separate leachables from burnables. Hood glove fingertips are cut off. The tips are routed to burning and the remainder of the gloves to leaching. All metal items are also directed toward leaching. Upon

completion of sorting, all items designated as burnable are placed in a large commercial chopper or hogger, and the material is reduced in size to approximately one-fourth inch. These pieces are then conveyed to a continuous belt incinerator which is electrically heated to 800° C. The material passes through the incinerator and is dropped free from the belt, as a plutonium ash, at the product end. The contents of one waste carton is reduced in volume to approximately two one-liter cans, representing a 98 percent volume reduction. After cooling, these cans of ash are neutron counted for plutonium value. They are then returned to the Feedstock Material Control Center to await further processing.

The material routed to leaching is handled in an adjoining hood where critically favorable pots are located. The material to be leached is given a continuous series of three separate rinses in a mixture of nitric and hydrofluoric acid. Upon completion of the third rinse, experience has shown that the items are free from recoverable amounts of plutonium. The rinse solutions are transferred from the three pots to an accumulator tank; mixed, volume determined, and sampled. These samples are analyzed, using standard chemical analytical techniques. The solutions are then piped to geometrically favorable cans. The plutonium solutions are transferred back to the Feedstock Material Control Center to await further processing.

Both the ash and liquid plutonium values are transferred to their respective categories in the EDP record system.

Solids residues are bagged from the hood and again placed in standard waste cartons. These waste cartons are neutron counted and, based on results obtained, are directed for either rework or burial--whichever is appropriate.

SOLIDS DISSOLUTION

The Solids Dissolution Material Control Center, the second of our processing operations, is shown on Chart IV. As depicted on our drawing, this control center consists of a series of glove boxes containing various types of batch dissolvers. The purpose in this operation is to reduce soluble plutonium-bearing scrap to a liquid state prior to transferring to the third processing operation, solvent extraction.

As the chart indicates, the first step is a reduction to the oxide form. This is accomplished by placing material for recovery into a hood and igniting to burn to oxide. Safety is the first consideration in this step, since the opportunity for receiving metallic particles in these types of scrap is always present. Failure to reduce these metallic particles to an oxide results in a pyrophoric hazard when the material is put in the dissolvers.

After assurance that no pyrophoric material is present, the next step is the actual dissolution.

For discussion, let us consider plutonium metal and plutonium oxide. As we have stated, we have purposely reduced these materials by burning. They are then transferred to one of the dissolution hoods, placed in a dissolver with a mixture of nitric and hydrofluoric acid, and, with heating, are allowed to percolate for a period of up to two hours. The solutions are vacuum-transferred to head tanks where they are measured for volume and sampled for plutonium content. Fresh acid is returned to the dissolver pot and the cycle is repeated. The number of subsequent cleanouts of the dissolver is based upon the analytical data obtained from sampling, combined with a visual check of the dissolver itself.

The solutions are then transferred to the Solvent Extraction Material Control Center, based on the analytical results and volume measurements from the accumulator tanks. This same procedure is used for the other feedstocks processed in this control center.

Solids removed from these batch dissolvers are neutron counted and, based on the plutonium present, are routed for additional dissolution activity, or are boxed for burial. The decision to either recycle or bury is based strictly on economic considerations.

In-plant material balances are maintained for each dissolver charge for criticality purposes. In addition, accountability records reflect a balance of each dissolver charge as it relates back to the feedstock input value.

SOLVENT EXTRACTION

Chart V covers the Solvent Extraction Material Control Center, the third of our processing operations. Plutonium solvent extraction is a process involving extracting and converting into a high-purity nitrate solution the plutonium content of the solutions from the other control centers or from currently generated liquid scrap from the other sections of the plant.

Two large continuous dissolvers are located in this section. They are used primarily for dissolving metal reduction and casting fragments, and are used infrequently for ash and metal alloy dissolution. Solutions from these dissolvers are combined with the liquids from the other control centers, and comprise the feed in this processing area.

Several feed accumulators are installed in this section. Feeds are being accumulated in one tank as the solvent-extraction system is being fed from another. Volumetric and analytical data are obtained prior to processing from a feed accumulator. These values are used as a control measurement to assure that acid and salt concentrations are within specifications for processing. In the solvent-extraction system, the plutonium is separated from the impurities, using tributyl phosphate; stripped from the organic; and then transferred to a product concentrator. The impurities follow the high-salt waste. The product is concentrated, measured, and sampled as official output from the plant. The high-salt waste stream is also measured and sampled, and then routed to an ion-exchange system to further reduce the plutonium content of this stream. Plutonium recovered at this stage is routed back to the feed accumulator. The high-salt waste is sampled, measured, and pumped to discard storage.

Sampling throughout these steps is used for both process control and accountability purposes.

UNIQUE PROCESSES

The last processing control center we call Unique Processes. It is shown on Chart VI. This phase of the operation is so named because of the array of material on hand that is not adaptable for direct input to the other processing centers.

We have shown two examples on the chart; there are others that fit in this category. The first example involves plutonium-chloride combinations. It is evident that, if scrap of this nature were introduced into the plant, the chlorides would have an adverse effect on the stainless steel equipment. Consequently, the chlorides must be removed prior to processing. This is achieved by reducing the scrap to a liquid state, precipitating the plutonium as an oxalate, and filtering the total charge. We have found that the filtrates are generally free of plutonium and can be discarded. The oxalates are removed from the filters and are transferred to the batch dissolver area in the Solids Dissolution Material Control Center for dissolution and subsequent solvent extraction.

The second example on the chart involves plutonium metallic alloys. The rate of dissolution of this material is limited by surface-to-liquid ratio; and we have found that by simple machining of these alloys to form machine chips we can increase this surface-to-liquid ratio and are able to achieve a fairly rapid dissolution rate. The chips are transferred to the continuous dissolvers in the Solvent Extraction Material Control Center for completion of processing.

HIGHLIGHTS OF ACCOUNTING, PHYSICAL INVENTORY, AND MEASUREMENT SYSTEMS

The daily activity in each of the processing areas is recorded by sequential run number on what is termed a "24-Hour Run Sheet." This form shows the identity and associated grams of plutonium charged into a particular processing run. The measured output is identified by analytical result, and the subsequent transfer point is designated.

The 24-Hour Run Sheets are certified by authorized personnel. They are forwarded daily to the Nuclear Materials Section, an independent administrative group that assembles and summarizes this information into an approved double-entry accounting system.

A complete physical inventory is taken monthly in the respective processing control centers. Normally, this inventory is taken with the processes shut down, tankage volume hold-ups reduced, and hood floors flushed. Transfers in and out are suspended until all inventory data and samples are completed. Strict inventory cut-off procedures are followed.

Inventory measurements are determined in the processing areas by the following: (1) Direct sampling and analysis of the process vessels and (2) Estimation of hood and hood floor hold-ups. (Physical inventorying by item control in the feedstock area has been previously described.) Certified copies of all physical inventories are forwarded monthly to the Nuclear Materials Section.

During the month, the Nuclear Materials Section maintains a running double-entry Book Inventory Balance for each control center. The recordings to the Journal Entry Work Sheets are made from the certified 24-Hour Run Sheets and other official transfer documents.

At the end of the month, the Book and Physical Inventory Balances are compared for reconciliation to the physical inventory.

A positive or negative "MUF" (Material Unaccounted For) is generated by the respective processing centers as a direct result of this reconciliation.

Statistical limits of measurement uncertainty are calculated on the inputs, outputs, and inventory, as operational control guidelines to compare with the reported MUF.

In the event that MUF exceeds these statistical limits, an analysis and investigation is started to determine potential causes. These causes are found in the areas of either unmeasured material or material incorrectly measured or recorded. The investigation covers the areas of potential gross measurement error, operating errors in sampling and inventorying, laboratory analytical errors, accounting errors, or inventory omissions. After the investigation, the causes are evaluated, adjustments are made, and operations are continued.

Formal reporting then emanates from the Nuclear Material Section, covering the total activity of the operation.

Even though measurement techniques contain uncertainties for individual plutonium measurements, over a long term the plutonium values have proven to be remarkably accurate. By applying suitable safety factors which reflect these

uncertainties, specifications for the safe storage, handling, and processing of scraps are developed. In addition, the overall inventory of various scrap types is known with reasonable certainty, since numerous individual measurements are used instead of single measurements in developing total inventory.

Although the present measurement methods prove adequate for Hanford "in-house" measurements, present methods may prove inadequate for the future. As plutonium fuel use increases, the number of shipper-receiver relationships on scrap shipments will increase markedly.

Present measurement techniques must be refined to handle the projected future volume and variety of plutonium-bearing scraps.

Hanford has gained experience over the years in the measurement of a wide variety of plutonium scrap types. Although appreciable effort has been expended and improved methods have been developed, the plutonium scrap measurement techniques available today contain measurement uncertainties, particularly when measuring low-plutonium-content scrap. This uncertainty, of course, reflects the difficulty of the measurement job itself--heterogeneous scraps are not simple to measure.

SUMMARY AND CONCLUSION

In summary, we have highlighted plutonium recovery from scraps at Hanford, from their sources to the final product.

The numerous items of plutonium-bearing scrap awaiting processing are stored in a critically safe manner. All items can be immediately located and identified for grouping and process matching.

During a normal month of operation, we handle approximately 4,000 items, representing several tons of solids which contain only a few kilograms of plutonium. It is important to note that the effort expended on items containing minor quantities of plutonium is as great as, and as complex as, the effort expended on items containing major quantities of plutonium. Items to be processed require scheduling of production runs through either the Incineration and Leaching, Unique Processes, Solids Dissolution, or Solvent Extraction

Material Control Centers. The recovered product, in the form of high-purity plutonium nitrate, is finally returned from the dormant scrap state to active use in the production stream.

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PLUTONIUM RECLAMATION OPERATION
MATERIAL CONTROL CENTER COMPOSITE

CHART I

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FEEDSTOCK MATERIAL CONTROL CENTER CATEGORIES OF Pu BEARING SCRAP 39FEEDSTOCK INVENTORY LOCATIONS 59CHART II

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INCINERATION & LEACHING MATERIAL CONTROL CENTER CHART III

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SOLIDS DISSOLUTION MATERIAL CONTROL CENTER CHART IV

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SOLVENT EXTRACTION MATERIAL CONTROL CENTER

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UNIQUE PROCESSES MATERIAL CONTROL CENTERINPUT EXAMPLES

Pu Al Pu Cl
Pu Zr

CHART VI