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SF Accountability Survey 17

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Survey #17

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Report on Survey of Accounting Control
Over Source and Fissionable Materials (DELETED VERSION)

University of California
Los Alamos Scientific Laboratory
Los Alamos, New Mexico

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Walter J. Williams, Director of Production — October 15, 1948
Washington
Walter C. Youngs, Jr., Chief, SP Materials Acct.
Branch, Division of Production, ORE, Oak Ridge
SURVEY #17 - SURVEY OF SP MATERIAL ACCOUNTING CONTROL, LOS ALAMOS SCIENTIFIC LABORATORY, LOS ALAMOS, NEW MEXICO (DECLASSIFIED VERSION)

REFER TO SYMBOL: MSP:WCY

Transmitted herewith are copies 1, 2 and 3 of the report of our survey of accounting control over source and fissionable materials handled by or in the custody of the Los Alamos Scientific Laboratory (University of California), Los Alamos, New Mexico.

The report was first placed in draft form and transmitted September 15, 1948, to Los Alamos for critical review and comment. Members of the SP Materials Accountability Branch (survey team) discussed the draft in detail on October 4, 5 and 6, 1948, with key Los Alamos AEC and contractor personnel, made various revisions, corrections, additions, or deletions as mutually determined to be desirable. It is believed that the present report properly reflects the status of SP materials accounting control at the Los Alamos Scientific Laboratory as of July 23, 1948, the date of the survey.

The Los Alamos Scientific Laboratory is in agreement with the findings as stated in the report. The major recommendations are acceptable to the Laboratory in principle and they have agreed to place them into effect as rapidly as the difficulties incident to personnel procurement, housing, working space, and equipment will permit. The Laboratory further agreed that although it is not feasible to place all recommendations into effect immediately, much can and will be done at once to integrate, improve, and give proper direction to the whole SP materials accounting program, as follows:

(1) The Laboratory will set up a central SP accounting control unit to handle all accounting for enriched uranium, plutonium, and normal uranium, integrating existing units into the central unit as feasible.

(2) The Laboratory will secure necessary additional people for the analytical laboratory so as to put the recommendations into effect on an orderly basis.

(3) The Laboratory will set up an adequate records system for enriched uranium, plutonium, and normal uranium.

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-2-

Walter J. Williams October 16, 1948

Walter C. Youngs, Jr.

SURVEY #17 - SURVEY OF SF MATERIAL ACCOUNTING CONTROL, LOS ALAMOS SCIENTIFIC LABORATORY, LOS ALAMOS, NEW MEXICO (DELETED)

REFER TO SYMBOL: WSF:WCY

(4) The Laboratory will provide for transfer data, inventory records, analytical data, etc., to feed into the central SF accounting control unit which will keep necessary records and relieve technical personnel of as much record keeping as possible.

The office of Santa Fe Operations, AEC, has agreed to lend all possible assistance to the Los Alamos Scientific Laboratory for placing the recommended program of SF materials accounting control into effect.

With respect to effecting the recommendations, the Laboratory accomplished a great deal between the completion of our survey on July 23 and our detailed discussions with key personnel on October 4 - 6, 1948. The most noteworthy accomplishment was the development in detail of a practical plan, ready for immediate application, for accounting control over plutonium, including records system, sampling, weighing, analytical procedures, internal material balance areas, etc. Another important accomplishment was the development of an accounting control plan for enriched uranium. This is an ideal plan which will be studied and discussed as a basis for devising a practical plan using present analytical procedures, equipment, and personnel. A great deal of commendable work has also been done in the analytical laboratory to improve and perfect procedures, particularly those subject to bias from various interferences (as described in our report).

In our opinion the work which has already been done, along with those recommendations which the Laboratory has agreed to place into effect at once, will do much toward establishing adequate SF material accounting control at Los Alamos. Once established, accounting control procedures should be improved and sharpened as rapidly as possible in accordance with the detailed recommendations in our report. The contractor should be encouraged and assisted in these endeavors in every possible way by SFO.

Walter C. Youngs, Jr.

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6 38 a

Survey #17

Survey of Accounting Control over Source and Fissionable Materials (DELETED VERSION)

University of California
Los Alamos Scientific Laboratory
Los Alamos, New Mexico

* Inventory note
See last 3 pages
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General Status

The recently issued AEC Bulletin GM-95 provides that each contractor handling SF materials shall be responsible for developing inventory and material balance procedures which will protect against undetected loss or diversion of SF materials.

We believe that the kind and quantity of SF materials at Los Alamos necessitates a more comprehensive system of accounting control than has yet been developed by the contractor, and recommend that an immediate effort be made toward this goal. This is our chief recommendation.

The three basic phases of adequate accounting control (records, measurements, and reliability) are not yet fully developed or effectively coordinated at Los Alamos. The records do not reflect some of the measurements which are made; many needed measurements are not made; and the reliability of the measurement data is not evaluated. A central unit, with full authority over and responsibility for developing and coordinating all three phases, has been found to be the most satisfactory means of establishing and maintaining adequate accounting control. Therefore, we recommend that such a unit be established.

Other major recommendations are that (1) all suitable measurements now being made for other purposes should be used for accounting control, (2) an improved plan of internal material balance areas should be established for each SF material, and (3) additional needed measurements should be made.

Many lesser recommendations and suggestions are contained in the body of this report.

In our opinion, the Los Alamos SF material accounting control system is incapable of assuring that all substantial amounts of SF material lost or misappropriated would be promptly disclosed, nor can it reliably show the SF content of the important quantities of material in residues. Therefore the Los Alamos Scientific Laboratory inventory and material balance reports cannot provide reliable information as to amounts unaccounted for and inventory totals.

It is a pleasure to record the friendly cooperation encountered during this survey.

Survey of Accounting Control over Source and Fissionable Materials

University of California
Los Alamos Scientific Laboratory
Los Alamos, New Mexico

1. A survey of accounting control maintained over source and fissionable materials at the Los Alamos Scientific Laboratory, Los Alamos, New Mexico, was conducted during the period July 7 - 23, 1948, inclusive. Participating in the survey were I. H. Hart, Jr., and W. E. Day of the Office of Santa Fe Directed Operations and a survey team consisting of W. C. Younge, Jr., D. E. George, H. W. Norton, and C. D. W. Thornton, of the Source and Fissionable Materials Accountability Branch, Division of Production, Washington, D. C.

Summary Description of Plant and Processes

2.1 Operations at Los Alamos are performed for the Atomic Energy Commission by the Los Alamos Scientific Laboratory (SFC) under a cost-plus-fixed-fee contract with the University of California. This contract is directly administered by the Office of Santa Fe Directed Operations (SFA).

2.2 These operations are chiefly the extremely important fabrication and assembly of weapon components, and include a program of research and development for improvement of weapons and of manufacturing processes. The Laboratory is also responsible for a continuing inspection and maintenance of weapons and weapon parts.

2.3 In the plutonium process, plutonium nitrate, $Pu(NO_3)_x$, is received from Hanford. This material is purified, converted to plutonium dioxide, PuO_2, and hydrofluorinated to plutonium tetrafluoride, PuF_4. The tetrafluoride is reduced to metal buttons, which are melted, cast, machined to proper weight, pressed, coated, tested and stored.

2.4 In the fabrication of fissionable parts from enriched uranium, green salt (UF_4), is received from Carbide and Carbon Chemicals Corporation, Y-12 Plant, Oak Ridge, or manufactured from residues accumulated from past processing. The green salt is then reduced to metal buttons. Similar to plutonium operations, several buttons are melted, cast, machined to proper dimensions, plated, tested, and then stored. Recently, buttons and machined castings have been received from the Y-12 Plant.

2.5 In the processing of normal uranium, scrap metal received from Hanford is melted, cast, machined and tested.

2.6 In each of the above processes, residues are accumulated, those from normal uranium being converted to impure black oxide (U_3O_8) and sent off the Area for recovery. The residues from processing the plutonium and enriched uranium are partly recovered by SFC; the unrecovered portions being stored.

Quantities of Material Handled

3.1 Major transfers of material to SFC for the first six months of 1948 were reported as:

Plutonium nitrate from General Electric Co., Hanford	59.6 kg Pu
Normal uranium metal scrap from General Electric Co., Hanford	17,057 kg U
Normal uranium metal parts from Carbide and Carbon Chemicals Corp. (Y-12 Plant)	595 kg U
Enriched uranium (about 93.5% U-235) from Carbide and Carbon Chemicals Corp. (Y-12 Plant)	
As green salt (UF_4)	210.8 kg U
As metal buttons	151.7 kg U

3.2 Major transfers of material from SFC for the first six months of 1948 were reported as:

Enriched uranium (about 95% U-235) to General Electric Research Laboratory, Schenectady, N. Y.	
As metal disks	46.9 kg U
Enriched uranium (about 94% U-235) to Research Division, Oak Ridge (for GE Research Laboratory)	
As metal disks	7.0 kg U

3.3 Transfers to and from Joint Task Force #7, and minor miscellaneous transfers involving several different consignees and consignors, are intentionally omitted from this list.

3.4 In kg of metal content, the plant inventory report by SFC at June 30, 1948, was:

	Normal Uranium	Enriched Uranium Uranium	Enriched Uranium U-235	Plutonium
Feed Storage	23,068.1	65.9	61.8	9.9
Material in Process	3,245.5	1,404.5*	1,320.6	55.5*
Material in Research and Development	22,030.8	31.0	24.7	40.9
Inactive Material **	5,497.6	316.9	297.3	33.3
Product - Parts inspected and passed	-	511.4	479.5	268.9
Total Inventory	53,842.2	2,329.7	2,183.9	408.5

** Inactive material includes material either not available or not intended for immediate processing. It includes all waste or scrap materials.

3.5 In addition, the reported inventory included 2.2 kg of depleted uranium, 77.9 kg of thorium, and 0.001 kg of U-233.

3.6 The inventory procedures underlying these reported quantities are discussed in the following sections of this survey report.

Material Unaccounted For

4.1 Material unaccounted for and material lost at SFC was reported, in kg of metal content, for the first six months of 1948, as:

	Material Unaccounted For	Known Losses
Normal uranium	4,322.4	173.8
Depleted uranium	None	0.2
Enriched uranium	None	*
Plutonium	None	None
Thorium	None	1.0
U-233	None	None

* For enriched uranium, a "known gain" of 16.8 gm (containing 15.5 gm U-235) was reported.

4.2 This unrealistic report that no material was unaccounted for in either plutonium or enriched uranium operations must arise from the accounting method in use, in which the difference between the amount of material at the beginning and that at the end of each process is assumed to be in the by-products.

4.3 However, the SF content of these by-products may be less than this calculated difference. Some losses, however small, are inherent in all production operations. Losses or misappropriation during processing

will not be revealed by this accounting method until the by-products are recovered. Because recovery methods have not been fully developed, disclosures of loss or misappropriation may be far in the future.

4.4 On the other hand, actual SF content of the by-products may be larger than shown by the records. In such instances, misappropriation of the amount present in excess of the book value would not be disclosed by the records. In addition, recovery processing may be delayed or omitted because the book value is too low.

Adequacy of Accounting Control

5.001 An approved AEC objective is the establishment of "a measurement and record system for transfers and inventories which will protect both the Atomic Energy Commission and its contractors against undetected loss or diversion of SF materials, and, in addition, will promptly provide current and accurate information as to the disposition and availability of SF materials". (Bulletin GM-95).

5.002 Although the kind, quantity, and form of fissionable materials combine to make the accounting control safeguard more urgently needed at Los Alamos than at any other Commission installation, SF material accounting control at Los Alamos is weak compared to that at the other major installations processing fissionable materials.

5.003 The two principal safeguards against loss or misappropriation of SF materials are physical protection and accounting control. Both are important, and neither is adequate alone.

5.004 Physical protection attempts to prevent loss, but gives no indication of the magnitude of losses which do occur. Accounting control is necessary to show what quantities of material are lost, and thereby to know how effectively the physical protection procedures are functioning.

5.005 Hence any weakness in accounting control over SF materials is a weakness in security.

5.006 The fissionable materials processed at Los Alamos are of extreme importance. Undetected misappropriation of these materials could seriously endanger the welfare of the United States. However, at this installation reliance has been placed primarily on elaborate physical safeguards and on the integrity of carefully screened personnel, without a system of material accounting control which would show promptly whether physical protection procedures are functioning effectively.

5.007 There are three basic phases of adequate accounting control over source and fissionable materials. Briefly, these are (1) a records phase, in which procedures are designed for recording and reporting interplant and intraplant transfers and plant inventories; (2) a measurement phase,

in which suitable measurement methods are designed to provide accurate determinations of the quantity of SF materials involved; and (3) a reliability phase, in which statistical methods are used to evaluate the accuracy and precision of measurements, that is, the reliability of the data.

5.008 Despite their dissimilarity, these component phases are closely interrelated. Their effective coordination is a requisite to achieving adequate accounting control. Data on inventories and material flows involving a number of different plants and departments must be integrated and comprehensively evaluated to ensure reliable totals. Technical data must be closely and continuously correlated with the records system. A central unit, with full authority over and responsibility for coordination and development of the three components (records, measurement and reliability) of source and fissionable materials accounting control, has been found to be the most satisfactory means of establishing adequate accounting control. Such a unit would have no control over production, but should be a valuable addition, improving and simplifying production records. Of course, the personnel of the central unit need not occupy a single office, but might advantageously be distributed through the plant, and should carry much of the bookkeeping burden now borne by production personnel. Location of the unit in the organization is a matter for administrative determination, but the requisite authority to get the information needed for accounting control should be provided.

5.009 The Los Alamos SF material accounting control system cannot assure that all substantial losses and misappropriations would be disclosed, because many quantities shown by the records are not ascertained by actual measurement. In particular, this is true of important quantities of SF materials shown by the records to be in residues.

5.010 Adequate material accounting control has not been achieved at Los Alamos in part because of the great emphasis on process development and on production, the nation-wide scarcity of qualified personnel, the difficulties of recruitment, the shortage of laboratory space and equipment, the housing shortage, and so on. To achieve adequate accounting control, the objective must first be clearly defined by management. Responsibility for its accomplishment must next be assigned to a person with both the time and ability to do the job, and he must be given the necessary backing, facilities and personnel. In general, the desirability of this safeguard has been recognized and acknowledged at Los Alamos. It has also been recognized that difficult measurement and inventory problems are always involved, requiring the efforts of expert scientific personnel.

5.011 Los Alamos personnel have considered some of the problems and have ideas for their solution. We expect this effort to result in continuing improvement to the existing accounting control system. The following

sections of this survey report discuss various aspects of the accounting control problem, some of which are peculiar to Los Alamos operations, and offer suggestions toward the development of a fully adequate system.

Records

5.101 Work sheets and supporting data for the June, 1948, inventory and material balance report were examined. A complete audit of the supporting records was not made.

5.102 In general, we feel that the records system is not sufficiently coordinated with material measurements to adequately reflect quantities of material received, shipped, on inventory or lost.

5.103 AEC Bulletin GM-95, issued July 23, 1948, prescribes minimum standards for SF material accounting record and reporting procedures. These requirements will necessitate the following changes, among others, in Los Alamos procedure:

a. Show material types on monthly inventory and material balance reports, with major types shown separately. (At present such information is partially shown for inventories.)

b. Report material balances for both the current month and the current year to date. (At present no material balance for the year to date is reported for enriched uranium.)

c. Include in the monthly reports comments as to basis of inventory.

d. Include in the monthly reports information as to the probable reliability of the various items. If such information is not available, that fact should be explicitly stated in the monthly report.

e. Use SF Shipping Forms to record transfers between SFC and other accountability stations.

5.104 The responsibility for the records of source and fissionable materials at SFC is divided into three independent sections. One section, Material Control, keeps records of plutonium. A second section, Quantity Control, keeps records of enriched uranium. These two sections also physically transfer these materials between divisions. A third section (referred to below as the "Tuballoy" section) keeps the records on normal and depleted uranium, thorium, and U-233. The first two sections are under the respective operating groups responsible for plutonium and enriched uranium fabrication, while the third section is in the Laboratory's property section, under the administrative division.

5.105 The Material Control Section is responsible for recording movements of plutonium only within the group responsible for the manufacture of weapon parts. When plutonium is transferred out of this group, it passes out of the jurisdiction of the Material Control Section. Further movements are recorded by personnel of the group having custody of the material. The Material Control Section has no control over such records, and can only maintain book inventories of the plutonium held by other groups. These book inventories are not amounts of material transferred to these other groups, either directly from the group to which the Material Control Section belongs, or between these other groups as reported by them to the Material Control Section.

5.106 In reviewing this system and its records, several specific items were noted and discussed with the Chief of the Material Control Section. The inventory reflected in their books at the time of the survey is not fully supported by inventory tally sheets. A physical inventory of the material within the plutonium operations group is made by visual inspection, and weighing in some instances, and is compared with the quantities recorded on the books. Containers of feed from Hanford and containers of inactive materials are visually inspected, the weights being assumed to be the book values. Plutonium in the form of metal is check-weighed at each inventory period. However, the inventory of plutonium in the possession of other organizational groups is not confirmed in writing to the Material Control Section. As this Section is responsible for the preparation of the plant plutonium material balance but has no jurisdiction over the preparation of reports by other groups having custody of plutonium, it appears that a written statement of inventory for each group signed by the group leader is a necessary minimum requirement.

5.107 The present arrangement of buildings in the plutonium area is carried into the design of the records system, that is, internal accounts have been established for each building. Such a method is satisfactory when sufficient measurements can be made to develop material balances around each building. When such measurements are not available, a more effective system can be developed by outlining material balance areas between process points where suitable measurements can be made. When all sidestreams are measured such a system permits the current disclosure of processing gains and losses. When all sidestreams cannot be measured, the system should be designed to have no more than one or two unmeasured sidestreams in any one material balance area. Some processing losses will then be disclosed upon the separate recovery of these sidestreams. Under the present system, where the plutonium content of all sidestreams are lumped by difference into a "Recovery" account, process losses or diversion will not be revealed until all plutonium is being recovered on a current basis. Action should be initiated to develop material balance areas around measurement points. This is further discussed in the section on "Measurements".

5.108 SFC has always used the Hanford measurement of the plutonium nitrate for material accounting. Present practice is to use this measurement of a batch as the initial debit, to subtract the weight of the button, and to transfer the difference to an account called "Recovery". Recently, some of these transfers have been minus quantities. This occurs when the button weight is greater than the Hanford value for the weight of plutonium in the batch. As discussed under "Measurements", we believe the initial debit to the records should be established by the SFC measurements. When the output of a process apparently exceeds the input, we believe it would be better to report the apparent gain than to record a negative quantity to be recovered.

5.109 Further, Hanford measurements have averaged higher than Los Alamos measurements. By June 30, 1948, the differences had accumulated to 2.802 kg Pu, this being reported as "Inactive Material". Because this quantity is not allocable to any particular lot of material, and because of uncertainties in the measurement methods, we believe that the weight and assay discrepancy account should be abandoned. The current balance should be reported as material unaccounted for. Future discrepancies should be reported as material unaccounted for or overaccounted for, as the case may be. If, upon completion of recovery processing of all plutonium residues the whole or any part of the quantity represented by assay discrepancies is found, the amount found should be reported as a gain.

5.110 Several minor points were also discussed with the Chief of the Material Control Section.

5.111 The records on enriched uranium are kept by the Quantity Control Section. The above comments on the plutonium records are also applicable to the enriched uranium records, that is, internal material balance records have not been set up on the basis of process points where suitable measurements can be made; the difference between the charge weight (shipper's measurement) and the weight of metal produced is charged to recovery; many of the detail records are kept by operating personnel rather than accounting personnel; and reported plant inventory is not fully supported by departmental work sheets.

5.112 The Quantity Control Section has the additional problem of accounting for the U-235 content of the uranium. Green salt or metal from Oak Ridge is received in batches of varying U-235 concentration. These batches are mixed in making a casting. The U-235 percentage for the mixture is then calculated from the batch concentrations and this calculated value is used for accounting control on the resulting casting and sidestreams. As these sidestreams are recovered and converted to green salt, a sample is sent to Oak Ridge for isotopic analysis, and the reported analysis is used for accounting control on the green salt. Variations between the calculated analysis and the reported analysis are accumulated algebraically in "Recovery". This practice has resulted in an apparent increase in the

U-235 content of the unrecovered material, the reported analyses of recovered material averaging lower than the calculated analyses. Reported analyses have averaged only about 93.5% as compared to 94.0% for the average calculated analysis. This corresponds to about 650 grams of U-235. This difference may result from a change in isotopic measurement methods or standards, from the accidental or intentional inclusion of some normal uranium in the enriched uranium recovery material, or from other causes, and it is believed that the most realistic procedure is to record and report the difference in U-235 as "Material Unaccounted For". When all the recovery material is finally processed, it may be that all the U-235 originally debited to recovery will be found. In such case, a gain would be reported.

5.113

5.114 Only a very cursory examination was made of the records kept by the Tuballoy Section. This Section is under the Contractor's Property Section and no chemical or physical advice or assistance has been used in the establishment of material balance areas or in the accurate presentation of the monthly inventory and material balance report. All detail records are kept by operating personnel and monthly inventory reports are furnished to the Tuballoy Section for consolidation into a plant report.

5.115 It was noted that no normal uranium was being reported as "Product". All completed parts were being reported as metal under "Feed Storage". Reporting "Product" as "Feed Storage" is not only in contradiction to existing regulations, but also results in the AEC consolidated SF report being incorrect.

5.116 The maintenance of all SF accounting records by a single accountability unit should eliminate many of the deficiencies noted above.

Measurements

5.201 The measurements phase of the accounting control system at Los Alamos was examined in detail. Meetings were arranged for us in which key operating and analytical personnel participated in detailed discussions of material balance areas, process points suitable for accounting control measurements, inventory methods, sampling and weighing procedures, and so on. Supervisory personnel also accompanied us during inspection of all plant operating areas, and fully explained past and present operations. Storage of feed, product metal and fabricated parts, residues, and intermediate materials was also inspected.

5.202 Our inquiries concerning methods available to verify inventory records of finished metal parts, which may not be sampled, were answered by scientists engaged in tests of nuclear properties of finished parts. Operations with the "water boiler" and with the "fast reactor" were observed and the scientists in charge described them for us. The analytical laboratories were inspected, including some examination of laboratory transfer and material balance records. All obtainable written analytical procedures were reviewed, and typical data were scrutinized.

5.203 Our opinion, based on this inspection, is that much remains to be done in the measurements phase, and that this is true partly because of the lack of facilities and the shortage of personnel. This opinion is based on such considerations as the desirability of improvements to various analytical procedures, the dearth of research on both analytical procedures and sampling methods, and the complete lack of facilities for isotopic measurements.

5.204 The existing internal material balance areas at Los Alamos are necessarily not fully effective, as discussed in the section on "Records". The subdivision of a complex plant or process into suitable material balance areas helps to localize losses and misappropriations by dividing the whole into manageable parts. To realize maximum benefits from such a sub-division, all the material inventories and material transfers in and out of each area must be adequately measured. The information produced will then be useful for locating trouble spots where improvements are needed in process, measurements, or physical security. The material balance areas in use at Los Alamos are not fully effective because too many of their input, output, and inventory items, cannot feasibly be measured.

5.205 We consider this a major weakness, and recommend that efforts be made to establish more suitable and effective internal material balance areas. These efforts should start with consideration of the whole of a production process, including all its attendant recycles and residues, to

determine the points where reliable measurements are feasible. Next an attempt should be made to develop and adopt a complete system of internal material balance areas which will make maximum use of these measuring points. To help to reveal clearly all significant abnormalities, statistical quality control procedures should be applied to the amounts lost or unaccounted for in each area.

5.206 At Los Alamos, as elsewhere, there are some material types for which measurement is impracticable. Such items hinder the attainment of a satisfactory and complete system of internal material balance areas. For some of these materials measurement is impracticable because the requisite analytical procedures have not been developed, or it may be that the difficulty is in sampling. The only practicable way to ascertain the SF content of such materials may be to process them for recovery, getting as much of the SF content as possible into a form that can be measured. When recovery processing is to be used, the segregation and separate processing of each material produced by operations in a particular material balance area and during a particular material balance period is necessary to obtain all the data needed for accounting control purposes. The uncertainties arising from lack of measurements for some items can be localized by a suitable choice of material balance areas. This choice should minimize the number of unmeasured items affecting each area. While valid methods of measurement for such an item are being developed, its supposed magnitude (calculated as the difference between measured debits and measured credits) should be subjected to statistical quality control procedures to help to reveal significant variations.

5.207 The available methods of gross measurement, sampling, analysis, and recovery processing, will determine which of the many possible subdivisions into internal material balance areas will be most effective. The advantages offered by new or improved measurement methods should be incorporated into the accounting control system as rapidly as possible. There should be a continuing effort to improve accounting control, including the possibility of revising the plan of internal material balance areas.

5.208 Many measurements which are made for production control, and some specifications measurements, could be adapted to the needs of accounting control. This illustrates the importance of close coordination of the records, measurements, and reliability phases. These measurements are described and considered, and needed measurements are suggested, in the following pages. Measurements are discussed first for plutonium, next for enriched uranium, and last for natural uranium.

5.209 Most of the plutonium measurements needed for accounting control are being made. An additional verification of the nature of the material is provided by its physical characteristics exhibited during machining, and by the phase-specification and similar tests. This is specially valuable for material for which only gross measurements are available. The final tests, of nuclear properties, will also reveal any substantial substitution of other material intended to conceal misappropriation of

plutonium. Aside from weights of pieces of plutonium metal, such as buttons and castings, the accounting control over plutonium stored or in process is not based on measurements of plutonium content. The residues, amounting to about 8% of the throughput if turnings be included, are evaluated only by difference between measured debits and measured credits to the process.

5.210 The amounts of plutonium received from the General Electric Company, Hanford, Washington, for processing at Los Alamos Scientific Laboratories, has always been uncertain because of discrepancies between shipper's and receiver's measurements. These discrepancies are briefly discussed in the section on "Statistics". Although basically the procedures used at the two sites to estimate plutonium are both potentiometric titrations, there are some essential differences. The Hanford shipping lot is not sampled by the shipper. Instead, a volumetric sample is taken from the transfer vessel out of which the tared shipping container is filled. After transferring the material to the container, it is concentrated and shipped. This concentration usually causes some precipitation, and the presence of the precipitate created a sampling problem for the receiver. Failure to recognize this problem was once a contributory cause of differences between Hanford and Los Alamos analyses.

5.211 Current procedures have apparently eliminated this difficulty, but substantial disagreements still occur on individual batches. These disagreements may be due partly to retention of a "heel" in the transfer vessel from which the shipping container is filled. Unless the solution in the transfer vessel were carefully mixed before sampling, the sample would fail to be representative of the solution transferred to the shipping container. Such sampling errors, if they occur, would tend to compensate as the results are totaled. However, as mentioned in the section on "Statistics", a small sample of the available data failed to give any suggestion of the occurrence of such compensating errors.

5.212 Plutonium determinations at Hanford involve a specific gravity measurement made by weighing a 100-lambda sample from the transfer vessel. The accuracy of measuring the 100-lambda volume is probably the limiting factor for the reliability of the Hanford analysis. During the Hanford survey we were shown data which seemed to demonstrate that this procedure can be very precise, but we have no data on its precision in routine use.

5.213 The differences between the results of the Hanford and Los Alamos analyses are discussed under "Statistics". Their magnitude warrants a critical re-examination of all potential sources of error, including those usually believed to be of minor importance. Of course, this investigation should be planned using statistical principles of the design of experiments. Various aspects of the Los Alamos procedure which should be re-examined are discussed below.

5.214 At Los Alamos a sample of convenient size is taken by production personnel and submitted to the analytical laboratory. It is withdrawn

from the shipping container after the contents have been diluted, weighed, and stirred. The laboratory immediately weighs out six portions suitable for titration, promptness being necessary because the concentration changes, water being decomposed by alpha particles emitted by the plutonium. Because the portions are weighed, the determination is on a weight basis, and this appears likely to be more reliable than the small-volume measurement involved in the Hanford determination. Two of these portions are used for iron determinations by orthophenanthrolene colorimetry. Since the iron is of the order of one percent, reliability at least as good as 10% is needed for iron if plutonium is to be measured within 0.1%. Though this procedure for iron was carefully checked during development, and though there is occasional recalibration, its reliability in routine use has not been evaluated.

5.215 There are important differences between the two installations in the reduction step prior to the titration. Complete reduction to trivalent plutonium is necessary for an accurate quantitative titration. Hanford accomplishes the reduction by means of titanous chloride, which apparently assures complete reduction to the trivalent state. This permits the use of the well-poised titanous-titanic potential system to ascertain the consumption of oxidant by the excess titanous chloride, and provides a potential jump which signals that plutonium has begun to be oxidized. The amount of titrant which is consumed between the titanous-titanic potential change and the change from trivalent to quadrivalent plutonium represents the plutonium content, after correction for blank and iron, provided no other interfering oxidations are occurring.

5.216 An amalgam reduction such as that now used at Los Alamos should be equally effective as a reductor. However, it lacks a convenient check point for the completeness of reduction, though visual inspection of the color of the sample is said to be effective. This requires the ability to distinguish that the final solution is a clear blue with no olive or greenish tinge. Routine verification that the potential of the reduced solution indicates complete reduction is recommended as an improvement to the Los Alamos procedure. This potential should become part of the record, showing that reduction was complete before the beginning of titration. Occasional spectrophotometric checks may also be helpful for ascertaining completeness of reduction, since the plutonium ions exhibit characteristic absorption peaks.

5.217 It appears desirable to monitor spectrographically the ion matrix of the Hanford material, in order to further insure the accuracy of the plutonium measurements. The cost of spectrographic analysis is low, in view of the information obtained, and the facilities are already available at Los Alamos. Such monitoring will help to prevent unrecognized interference by other ions which are reduced by mercury amalgam and are titrated by ceric ion as if they were plutonium. This is particularly important when an arbitrary potential is taken as the end point, or where other reduced ions are undetectable, because the method contains the assumption that the oxidant is consumed only by plutonium and iron.

5.218 At present Los Alamos titrates a weighed sample, the reliability of

weighing being superior to the reliabilities of other measurements involved in the determination. However, the endpoint of the titration is calculated, assuming it to correspond to an arbitrary potential of 760 millivolts, rather than being determined in the classical fashion, wherein the endpoint is taken to correspond to the maximum in the rate of change of potential per unit of titrant. Such a departure from common practice is warranted only if substantial experience shows that all the electrode systems used change at the same potential. The errors due to such variations and the ones discussed below are possibly insignificant since a ten millivolt change is produced by only 0.2 milligram of titrant. As a precaution, however, it would be desirable to make a potentiometric plot for each electrode system, at the time of fabrication or resensitization, to ascertain the break point, that is, the potential at which the rate of change is greatest. Such a precaution is desirable because temporal changes in the potential of reference electrodes are common, and because different indicating electrodes may break at slightly different potentials. Since the meter-read potential is measured against a reference electrode, changes in that electrode would result in biases in the measured potential. Also, biases may result from the practice of calculating the endpoint from an arbitrary potential. Any bias in the endpoint potential would cause a bias in the amount of plutonium reported.

5.219 The Los Alamos accounting control system now uses the amounts of plutonium determined by the General Electric Company at Hanford. However, it is usually desirable to base the material balance of a plant entirely on measurements made at that plant. Biases in measurements of the input, by-product, and output streams will then often be similar and tend to cancel. If sufficient precautions are maintained, the analytical procedure in use at Los Alamos should yield satisfactory measurements, and it is recommended that Los Alamos use its own measurements for material accounting control.

5.220 The first step in purifying the material received from Hanford is reduction to trivalent plutonium, using hydriodic acid, followed by precipitation as the oxalate. The so-called "oxalate filtrate" and washings from the shipping containers are combined, and are concentrated and stored for future recovery. Measurements of the plutonium content of this concentrate are being made but are not used in the accounting control program. It is recommended that these measurements be used for accounting control records. When the plutonium contained in the filtrate concentrate is recovered, differences between amounts debited and amounts actually recovered will appear as gains or losses.

5.221 The oxalate-filtrate plutonium content is measured by radioassay. For this technique to be useful for accounting control, attention must be given to certain aspects of the present procedure which have heretofore been of minor importance to the production personnel.

5.222 The radioassay method should be based on experimental data for corresponding Hanford product. The calibration curves now used for the radioassay method are based on pure plutonium which was produced at Hanford in the early days of operations. Satisfactory use of the

radioassay method for accounting control is dependent upon knowledge of variations in the specific alpha activity of the plutonium isotope mixture currently being handled in the plant. For both operating and material balance purposes, it appears desirable to experimentally strengthen the foundations of the radioassay method by continually utilizing plutonium metal-plant product for counting standardization. The laboratory supervision has been on guard against changes in Hanford pile operating levels, which will probably affect the specific alpha activity of the product and hence would require recalibration.

5.223 Statistical quality control should be continued on the accuracy and precision of the counting methods, and the inherent statistical counting error should be reported.

5.224 The correction for internal absorption should be periodically reviewed while it continues to be used, and the reported values should be corrected for this absorption as discussed below in the section "Statistics". A rough estimate of solids content is submitted with each sample to aid estimation of internal absorption corrections. This has proven unsatisfactory, and we believe that the possibility of chemical purifications prior to counting should be carefully reconsidered with the object of including such purifications in the counting routine.

5.225 Radioassays corrected for internal absorption should be sufficiently accurate for accounting control purposes since the oxalate filtrate contains only about 0.1% of the input stream. Errors in these measurements usually can be easily kept as low as 10%.

5.226

5.227 The spectrographic monitoring of Hanford product shipments discussed above may serve a further useful purpose by enabling correction of the plutonium content of the plutonium dioxide obtained by use of a conversion factor, the correction being for the lanthanum content. The quantitative determination of lanthanum in the respective Hanford batches would enable the dioxide weight to be corrected if lanthanum oxalate is quantitatively precipitated or co-precipitated with the plutonium oxalate. A spectrographic or chemical method should be investigated and the distribution of lanthanum to the several process streams should then be studied.

5.228 In the following discussion, examples of internal material balance areas which appear to us to be appropriate are suggested and briefly described. These suggested areas have been indicated on the attached

flow diagram for the plutonium process, which will therefore be helpful in following the discussion. It should be remembered that these suggestions are quite tentative and incomplete, and are restricted to the limits currently imposed by Los Alamos measurement methods and procedures. A complete plan is needed for plutonium, for enriched uranium, and for natural uranium. In developing these complete plans, Los Alamos personnel who are fully acquainted with their processes and problems will probably be able to improve greatly on these suggestions.

5.229 Because the efficiency of hydrofluorination is variable, the first suggested internal material balance area for the plutonium process begins with the material received from Hanford, and closes with the conversion to the dioxide. The principal debit to this area would be the plutonium content of the Hanford nitrate, as determined at SFC. Other debits would be made when plutonium from the recovery process is fed into the production line. The chief credit would be the plutonium content of the dioxide, with two minor credits for the plutonium contents of the oxalate filtrate concentrate and of the nitrate sample. As all of these flows are now being measured, the amount of plutonium lost or misappropriated in this area could be determined by such a material balance. This amount would include any dusting in the conversion from oxalate to dioxide.

5.230 The second suggested material balance area includes hydrofluorination of the oxide and reduction to metal. The debits to this area would be the amounts of plutonium in the dioxide fed to hydrofluorination and that in turnings recycled to the reduction process. The chief credit would be plutonium contained in the metal buttons produced, with minor credits for that in crucible, slag, and button pickling solutions. The amount of plutonium contained in crucibles and that in slag is not being measured. Until these materials are analyzed, the material balance for this area will remain incomplete. The difference between measured debits and credits may be calculated, and may be attached to the unmeasured amounts as an indication of what should be found when they are measured or processed for recovery. Until then, however, there is no way of ascertaining the amount lost or misappropriated for this area. It should be noted that there is a presumably small but unmeasured amount of dusting loss from the hydrofluorination process.

5.231

5.232 The excess hydrogen fluoride gas used in the conversion and the gasses produced are vented to the sewer by way of a water aspirator. Although dusting tendencies are slight, losses may occur in this operation. Such losses as do occur are not detected, and would be known only if the sewer is continuously sampled and analyzed. Such a procedure is feasible at other plants, and because of the exceedingly high alpha activity of plutonium, which enables easy detection, we recommend

continual checking of the sewage until evidence is accumulated that losses are small and infrequent, followed by occasional checking. In another plant, engaged in hydrofluorinating uranium oxide, it was found that losses by entrainment in the gas sweep were high enough to necessitate recovery of the entrained material. Because sewer measurement is complicated by the large aspirator flow, chemical absorption of the sweeping gas in a trap, the contents of which are evaluated, may be a preferable method.

5.233 The typical reduction charge consists of plutonium tetrafluoride and turnings of plutonium-gallium alloy, with iodine and finely divided calcuim. The plutonium buttons produced are pickled in nitric acid and weighed, and the pickling solutions are stored. We recommend that these solutions be analyzed. The allocation of plutonium to slag and crucibles by difference would thus be considerably sharpened.

5.234 The turnings are sampled and analyzed for gallium content as a specifications measurements for finished metal parts. These analyses suffer some inaccuracy because of segregation of gallium in the alloy. Attempts should be made to determine the amount of gallium in buttons produced from charges that include gallium-containing turnings. Some of the technical personnel stated the opinion that nearly all the gallium in the charge turnings appears in the slag. The weight of plutonium inferred from the button weight should take proper account of the fate of this gallium. (Proper allowance for the alloyed gallium should be understood throughout the remainder of this discussion.)

5.235 It is desirable to know whether the gallium added at this stage is all present in the rough casting produced. Such information may be obtained from the gallium measurements now being made on the turnings, provided marked segregation has not occurred near the surface turned, or perhaps could be ascertained by a chemical examination of the "skull" produced. It was felt by the operating personnel that the weight of the skull proper, which can be broken from the pouring crucible, would differ seriously and erratically from the weight of plutonium in the skull and so would have little significance for accounting control. These skulls are now being recycled into the purification step of the process as a solution of the skulls in hydriodic acid.

5.236 Two means of handling this recycle in the accounting control program are available. One would involve a separate processing of skull solution through the purification process to produce plutonium dioxide which would be weighed. The plutonium content found at this point would be debited to the purification operations and would be credited to the casting operation. This method is unattractive because losses in the purification operations would not be detected nor allowed for. The alternative procedure is to analyze the skull solutions, with such

compositing as may be suitable. Although the analytical techniques involved in the evaluation of this stream may be not entirely satisfactory at present, the concentration level is sufficiently high, and the impurity matrix should be sufficiently low, that wholly satisfactory analytical techniques could be developed. The casting crucible account would probably have to be a difference item but would be separate and better defined than if the skulls were unmeasured.

5.237 The fourth suggested area includes only machining of castings. This area is debited with the weights of castings. If the presence of significant amounts of oxide or other impurities on the surfaces of the castings is suspected, the plutonium content of turnings should be determined occasionally. It will probably be related to the size of the casting from which the turnings come and to the thickness of material removed by turning. Credits would include plutonium content of machined parts, samples, and turnings, machining dusts being temporarily evaluated by difference. Upon processing dusts or dirty turnings in hydriodic acid, they would be handled in the same manner as the "skulls". It may be necessary to increase or improve present knowledge of the plutonium content of the turnings. Since samples are already being taken for the gallium specifications analysis, it may be that no further sample will be needed. However, because of the sampling difficulty attendant upon segregation, a series of plant experiments may be useful. Thus batches of turnings could be accumulated and carefully processed separately to plutonium dioxide so as to ascertain their plutonium content. Except for the small losses during processing, the plutonium content of the dioxide would be equal to that of the turnings processed.

5.238 The fifth suggested plutonium process material balance area includes pressing, cleaning, and plating. Weights of machined castings would be debited, and credits would include weights of clean parts ready for plating and amounts of plutonium recovered from flash and dust. Emery backing would be evaluated by difference. It should be clearly understood that these five suggested material balance areas are only examples to indicate possible improvements over areas now in use. They should not be considered to be either the only possible satisfactory areas or the only ones needed. A complete system, covering all aspects of the process, is needed. In developing a complete system it may be necessary or desirable to rearrange or further subdivide the areas we have suggested.

5.239 So far as practicable, each recovery material should be separately accumulated by monthly material balance periods and separately processed for recovery, or to a state suitable for measurement. Real gains or losses will then be revealed by comparison between the amount recovered and the amount previously debited for this particular material. Upon completion of recovery of the plutonium contained in any particular residue accumulated during a particular material balance period, the gain or loss should be transferred to a separate gain and loss account and should be reported as material unaccounted for.

5.240 After the pressed parts are cleaned they are plated by deposition of nickel by thermal decomposition of gaseous nickel carbonyl. It is believed that no plutonium is lost in the plating process. Plated parts are now being weighed, and it is felt that accurate weights should be recorded to enable future weight checking against substitution or diversion. Plated plutonium parts are subjected to a radiographic test for defects.

5.241 Parts rejected for faulty plating, or by later inspections and tests, are stripped with fuming nitric acid and replated. The stripping solution is being analyzed and these analyses should be used for accounting control.

5.242 The finished parts are transferred to M Division. Mock-up tests involving the nuclear properties are performed in the Pajarito laboratories, and substitution of another metal for the plutonium would be detected. Only such tests, involving neutron multiplication, are effective throughout the volume of the piece. They are not subject to the limitations of the fission-counting or alpha-counting tests, which test only the surface material.

"not" part in pencil memo 10/25/48. to W.J. Willis for MCY copy in Central Records

5.243 Periodic checking of fabricated fissionable parts will be a problem of increasing importance to the Commission as stockpiles of such units are accumulated and as transfers of such materials between installations become more frequent. The periodic verification of fissionable inventories as an accounting control problem and the mock-up testing of parts for weapon production are essentially similar. Research workers at Los Alamos should be made aware of this similarity so that information helpful toward the solution of the common problem will be effectively used. Focusing the attention of competent nuclear engineers on accounting control problems may lead to the development of simple instruments for determining whether parts coated with fissionable materials have been substituted for bonafide fissionable parts.

5.244 Over 18 kilograms of plutonium, in 41 steel-canned slugs, is being used in experiments with the so-called "fast reactor". These experiments are sensitive to a single slug and the unauthorized removal of a slug from the reactor would thereby be detected. The plutonium slugs not actually in use, together with a number of externally similar normal uranium slugs, with which they might be confused, are stored in a vault. Access to this vault is limited to a few persons. A convenient way of differentiating between the plutonium and uranium slugs is by slug temperature, the difference being noticeable to the touch. The energy of the alpha particles emitted by the plutonium appears as heat when the particles are absorbed by the steel can, maintaining the plutonium slugs at a temperature higher than that of the uranium slugs. The slugs may also be identifiable by precise weighing. It is recommended that the plutonium slugs be counted and infrequently weighed. Also,

because the individual slugs do not have distinct nuclear properties, they should be rotated in the experimental program so that a physical check of the nuclear properties of every slug will occur at least once every three months.

5.245 In addition to the slug inventory, the fast-reactor vault had an inventory of 461 grams of uranium containing 95% U-235, consisting of small blocks of uncoated metal, weighing a few grams each. They are to be used in experiments lasting about one year. Some of these blocks could be stolen, with or without substitution of similar blocks of normal uranium. In either case discovery would be delayed because the blocks were individually packed in small cotton-filled cardboard boxes and because the experimental program would never call for all the blocks at once. It is recommended that the cubes be physically inventoried, including weighing at least every three months, and that nuclear or other monitoring to check the U-235 content should be equally frequent.

5.246 The various vaults for storing SF materials need to be reorganized, largely because of the shortage of vault space. The plutonium recovery program has lagged far behind current production, and it has become necessary to store a considerable amount of miscellaneous plutoniferous materials in a locked room instead of in a vault. This is not particularly objectionable so long as no satisfactory recovery process is available.

5.247 The most important residues from plutonium processing are metal turnings and skulls. These are recovered by recasting the turnings and returning the skulls to the purification process. Plutonium recovery research has been generally aimed at the residues most likely to repay study, and this effort might be improved by detailed inspection of the plutonium inventory record and storage areas. A motley collection of analytical residues, accumulated during early days, remains to be processed. An important point in planning research in analytical methods for plants handling material of such high value is to attempt to find methods which will yield the valuable material at the end of the analytical procedures in a form suitable for feeding into some part of the production process, thus facilitating recovery of the analytical residues.

5.248 Considerable vault space may be gained by removing from the vaults such materials as the casting crucibles and various slags from reduction processes, for which no practical recovery process is known. Storage in the same vaults of any other material neither fissionable nor used in handling highly purified fissionable material should be discouraged. Empty Hanford shipping containers should not be stored in the same vault with full containers, because one of the simplest methods of purloining materials from plants is to include full containers in shipments of empty containers. Once outside the plant

limits there is less difficulty in stealing the contents of the full containers. Therefore, the storage and shipping depot for empty containers should be geographically well separated from the receiving depot for any valuable material.

5.249 Chemists and physicists should participate in developing improved procedures for checking vault contents. The utilization of fission or other counting devices at the vaults, the installation of scales and balances, and the systematic arrangement of vault contents to facilitate periodic inventories are major points, and should be reconsidered when additional vault space becomes available.

5.250 All enriched uranium now being shipped into this plant is in finished or semi-finished metal parts. Formerly the plant was supplied with green salt, uranium tetrafluoride, which was analyzed chemically and assayed isotopically at Oak Ridge, the total book inventory of enriched uranium for the plant being on the basis of shipper's figures.

This department is now concerned primarily with the recovery of material contained in skulls from the reduction process, turnings from parts fabrication, rags and similar materials, peroxide filtrates from turnings and skulls processing, and with liner material from the reduction operations. Present accounting control procedures credit the recovery account with the output from the recovery department, but the original debit for the material recovered always includes other materials which are not being recovered or measured. Therefore, it is impossible to know what processing losses are occurring or have occurred in the past until all of the accumulated residues are processed. It is recommended that new material balance areas be established and that the recovery materials be separately accumulated by material balance periods and material balance areas. Current processing losses could then be ascertained.

5.251 Skulls and turnings are oxidized and dissolved in nitric acid, and the uranium is purified by precipitation as the peroxide and conversion to black oxide, U_3O_8. We recommend that the nitric acid solutions be diluted to a standard volume or weighed, and sampled for analysis. These samples may be composited to decrease the analytical load provided suitable compositing controls are established. Analysis of these pure uranium nitrate solutions, although representing additional work of a novel kind for the laboratories, should not be difficult after a period of experience has been gained, satisfactory procedures being already available. A similar analysis, but requiring somewhat less accuracy, may be run on acid leachings from ashes of rags and towels. Plating and pickling solutions destined for storage should also be analyzed. The black oxide produced is weighed and this weight

should be used for accounting control purposes. This oxide is probably pure enough to justify use of an empirical factor for uranium content. Spectrographic purity controls are run which insure that the desired purity is achieved.

5.252 Uranium peroxide is separated from the supernatant solution using a fritted pyrex glass filter, the supernatant being pulled through the filter by suction. Between the vacuum pump and the filter flask there is a trap containing an ordinary inhalator mask filter, a screen, and powdered calcium oxide. It was stated that no significant amounts of uranium had ever been found on the trap material. The analytical methods used were not described. However, lucite hoods have been shown by health studies to be necessary for these operations, appreciable alpha activity having been detected in the air, on benches, and so on, before the hoods were used. It appears possible for entrainment to occur in the suction filtration, and the traps were designed to prevent such losses. Analysis of trap contents disclosed trifling amounts of uranium not worth recovery processing, but no tests have been made of trap efficiency. Because of the geometry of the filter, there is a possibility that the gasses by-pass the packing material. Failure to find appreciable amounts of uranium on the filter material is incomplete as a check on losses until it is shown that the filters are effective.

5.253 The peroxide filtrates are sent to M Building extraction plant for recovery processing. They are concentrated and analyzed prior to being fed to the extraction column. These analytical results should be used to credit the peroxide recovery operations and to debit the extraction plant.

5.254 The uranium content of the green salt produced in the hydrofluorination process is taken to be uranium content of the black oxide charged to the process. However, experience at other plants indicates that uranium tetrafluoride is an almost stoichiometric and stable compound, so that its uranium content may be calculated by means of a factor and it can be transferred on a weight basis. If this is not true at Los Alamos, the present procedure may have to be continued. Is not true!

5.255 Samples of green salt produced in recovery operations are sent to Oak Ridge for isotopic assay. The interval between sampling and receipt of the assay by Los Alamos is about two months, due chiefly to delays in transportation, the assay itself requiring at most only a few days. Though the assays are made solely for operating purposes, they would also reveal any substantial dilution, with normal uranium, which may have occurred.

5.256 A considerable inventory of green salt from recovery operations has accumulated. It is stored in a vault in unsealed glass jars with screw tops. Green salt entirely similar in appearance, and of identical chemical properties, which has been produced from normal uranium, is easily available to many plant personnel. Therefore, there is an important risk of substitution during the extended period between the time of production of the green salt and the time it is reduced to metal.

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5.257 The recovery program for liners from enriched uranium reduction and casting is essentially a solvent extraction and is still in pilot-plant operation. Attempts have been made to establish an engineering material balance around this operation, but have been unsuccessful for the few runs so far completed. This may be largely due to analytical difficulties which are discussed below.

5.258

5.259 The nitric acid concentrates have proved to be too impure to feed into the peroxide precipitation recovery process, and it is expected that further extraction purification may be necessary before these materials can be recovered. These concentrates are currently being analyzed, and these analyses and other extraction plant data should be used for accounting control. An adequate verification procedure for the inventory of stored material should be instituted.

5.260 Two major types of chemical analyses are being employed on solutions processed in or produced by ether-extraction recovery operations. A thiocyanate colorimetric method is used for intermediate amounts of uranium, and a liquid fluorescence method is used for trace amounts. The thiocyanate method is essentially that developed by Nelson and Hume, with modifications in the preparation of the sample. These modifications may need further investigation of their suitability.

5.261 Column charge samples are prepared for analysis by room-temperature precipitation of uranium and some other cations by means of ammonium hydroxide, iron being added if needed. The precipitate is then dissolved for thiocyanate colorimetry. Thus the method as used is not proofed against sporadic or continual interference by undecomposed hydrogen peroxide from the peroxide concentrates included in the charge, nor against interference by carbonate, which may have survived or may have been present in the ammonium hydroxide. These interferences would both tend to give low results due to soluble complexing of uranium in alkaline solution.

5.262 The color intensity which is finally measured changes with the passage of time. Therefore, routine check samples and qualtity control procedures should be used. Simple checks on precision alone are inadequate for procedures which are subject to such changes. Some checks have been made by spiking a few samples of process solutions. However, ordinary spiking techniques do not check against threshold interferences, and check against

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sporadic interferences only in the samples actually spiked. Fluctuations in the extraction plant material balances suggest that further study of the analytical methods is needed.

5.263 Because of the difficulties often encountered in the decomposition of peroxides in chemical solutions, it is recommended that some simple test for peroxide be instituted prior to the ammonium hydroxide precipitation, so long as this precipitation continues to be used. Of course, the whole method should be compared to independent techniques.

5.264 The fluorescence technique is extremely sensitive and very susceptible to interference. Therefore, it should be thoroughly checked with the solutions for which it is being used. It is applied to samples of the raffinate resulting from ether-extraction of recovery charge. However, the preparative treatment consists of analytical re-extraction of the sample with diethyl ether. The major weakness of such a procedure is that anything which makes the uranium non-extractable in the extractions column may also render it non-extractable in the preparative treatment. If so, the uranium would not be delivered to the solutions used for the fluorometric measurement. Hence it would be undetected, and the raffinate would seem so low in SF content as to justify discarding it.

5.265 Approximately 870 grams of U-235, contained in about 13½ liters of solution, constitutes the active material for the so-called "water boiler". This solution is contained in a stainless steel flask shielded by five feet of concrete. Most of the fission products are removed from the chamber by an air sweep. The sweep also removes hydrogen and oxygen produced by fission-fragment electrolysis of water. The sweeping and cooling arrangements provide a means of access to the fissionable material. However, the criticality of the mass is strongly dependent on the amount of fissionable material present, and the scientists in charge felt that the continuing experimental program would detect a diversion of only one or two percent. Because the facilities are inadequate for analyzing material which contains fission products, the operating characteristics of the water boiler appear to constitute the only practical verification of the inventory of this fissionable material.

5.266 The normal uranium shielding around the energizing element of the fast reactor should continue to be carried on the inventory for the installation, though some of the scientists suggested that this material be written off. Occasional inspection would disclose tampering and should be adequate inventory verification.

5.267 The present upper limit for discarding materials from the U-235 recovery operations is 0.1 ppm. Since the plutonium recovery operations are not yet a part of production, no plutoniferous raffinates are being discarded, but upper limit has been set on the basis of health requirements. It is important to note that there are sinks or washing facilities in practically all of the areas in which fissionable materials are handled. Although normal operating procedures are designed to avoid loss of material by way

of the sewer systems, there is no routine test for such losses. The delivery of material into the sewers could be either accidental or intentional. Such losses would probably occur only in spurts of solution of high SF content, and thus would be unlikely to be detected by intermittent sampling of the sewer contents. It has been found both practical and desirable at an installation handling only normal uranium to sample sewage continuously and to analyze daily. It would appear more desirable or even necessary that the major sewers at Los Alamos be continuously sampled and analyzed so that this possibility may be eliminated as an unknown in over-all material accounting control.

5.268 There are relatively few chemical measurements made on natural uranium for accounting control. The present material fed to the plant is largely scrap uranium metal from other Commission installations. It is debited to the plant on a weight basis as 100 percent uranium metal, although varying degrees of oxidation have occurred, and extraneous material is sometimes present. The scrap metal is remelted and cast, and the castings are transferred to the machine shop. Skulls from the pouring crucible, oxide burned out of the crucible, and material recovered from various stages in the processing, are burned to oxide, accumulated in small drums, sampled and analyzed, and shipped to other installations for recovery. Analytical values have not been used to credit the plant with materials shipped. Instead, an arbitrary factor of 0.75 was used up to the time of this survey. The problem of sampling the oxide has received some consideration. It should be solved, and analytical values for the oxide should be used for accounting control.

5.269 A commendable effort has been made to attain accurate accounting in the machining and fabrication operations. It is recommended that the proposed central accounting control unit institute studies of the handling of turnings so that experience factors may be found. These factors can be applied to turnings weights and should result in more accurate transfer accounts. A study of the compositing possibilities should be instituted to minimize the increase in analytical work, and every shipment of oxide from the plant should be evaluated.

5.270 A rudimentary material balance for all SF materials is kept by the analytical laboratories which, suitably modified, could become part of a system administered by a central accounting control unit. Improved analytical transfer and report forms were discussed with the laboratory personnel. More complete laboratory debits would result and, combined with evaluations of the salvaged analytical residues, would enable stricter accounting.

5.271 Few analytical personnel are committed to inventory and material balance measurements at this installation considering the value of the material being handled. We believe increased measurement capacity and improved and expanded measurement facilities are urgently needed if adequate accounting control is to be achieved. There should be a thorough survey of the measurement needs of the production operations, concerned chiefly with the measurements needed for better material balances and for better production control.

5.272 In addition to such a review, the uranium analytical procedures should be carefully written up, the plutonium procedures should be revised, listings should be prepared of the specific plant samples to which the procedures are applicable, and the analytical research needed to provide the comprehensive measurement system required for adequate accounting control should be determined. An important aspect of the analytical research is to develop measurements procedures for which residues of fissionable material may be returned directly to existing plant recovery cycles.

5.273 The lack of isotopic assay facilities at an installation handling various enrichments of U-235 results in a vulnerability to substitution or error which is unwarranted when processing material of such great value. The present metal-casting operations in Sigma building handle both natural and enriched uranium. The mixture of natural uranium with a considerable amount of weapon-level material anywhere in the plant would be a mistake costly enough to justify the expense of facilities for isotopic measurements. If mass spectrometric assay facilities cannot be obtained, the somewhat less satisfactory but still useful isotopic assay methods of fission counting and emission spectrography should be considered. However, there are operating spectrometer laboratories at other installations where personnel could be trained and from which some equipment should be obtainable for installation at Los Alamos. Wide dispersal of such equipment and skill, now localized at Oak Ridge, is desirable as a precaution against disaster and sabotage.

5.274 It should be pointed out that one of the chief advantages of an integrated accounting control system is the continual cross-checking of techniques and measurements. This helps to show which development and research efforts will pay the largest dividends in improvements to process, measurements, and accounting control.

Statistics

5.31 The statistical phase of accounting control includes evaluation of the precisions and biases of all the various measurements underlying inventory and material balance reports, and the proper combination of these precisions and biases to yield the probable limits of error for the amounts reported. Also, statistical principles should be used to ascertain the distribution of available effort which will be most effective in narrowing limits of error.

5.32 At present, there is no organized statistical phase of accounting control at this installations. Statistical activities as a phase of accounting control should be undertaken, and should constitute an integral part of the central accounting control unit proposed in this report.

5.33 Average differences of the order of ½% have always existed between Los Alamos and Hanford determinations of plutonium. Differences as large as 5% occur on individual Hanford batches, though the difference is as small as 1% on about half the batches. These differences have shown occasional

unexplained trends and sudden changes of variability, and at present there is no authentic explanation of the discrepancies either individually or on the average. The differences on individual batches should be subjected to a system of statistical quality control, with the object of revealing changes in either the average value or the variability. Causes can then be sought promptly, accidental improvements being incorporated when recognized, and additional precautions being taken against recurrence of adverse changes. In addition, a research project should be undertaken to investigate the nature of the difference, involving cooperation between Los Alamos and Hanford. Statistical principles of experimental design should be used in planning this project.

5.34 A possible source of difference between the Hanford and Los Alamos analyses is the heel retained in the transfer vessel from which the Hanford shipping container is filled. As mentioned in the section on "Measurements", failure to mix thoroughly the heel with the rest of the contents of the transfer vessel before withdrawal of the Hanford sample would cause the sample to be unrepresentative of the solution transferred to the shipping container. If such sampling errors occur, they would tend to be of opposite sign for successive batches. This would be manifested by a negative correlation among successive batch differences between Hanford and Los Alamos. Seeking evidence on the existence of such errors, we examined 510 consecutive batch differences. To avoid or minimize the effects of trends and changes in bias, the circular serial correlation coefficient was calculated for each of 51 groups of ten consecutive differences. If the supposed effect is nonexistent, the expected value of these correlations is -0.111. If the effect exists, the expected value would be more negative, depending on the magnitude of the effect. Of the 51 calculated correlations, only nine were as negative as -0.111. Moreover, none was as negative as -0.564, the value below which one out of 20 should fallpurely as the result of sampling when the effect is nonexistent. Therefore the data examined fail to support the hypothesis that incomplete mixture of a heel retained in the Hanford transfer vessel is contributing to the differences between Hanford and Los Alamos plutonium determinations. Of course, it may be that there is such an effect and that it is being overwhelmed by some other effect which is of opposite sign. In fact, six of the 51 correlations exceed 0.360, which should be exceeded by only one out of 20 if there is no effect, and two exceed 0.525, which should be exceeded by only one out of 100 if there is no effect. Therefore the data give substantial evidence that some factor is at work which causes similar errors in successive batch differences between Hanford and Los Alamos.

5.35 Some statistical work is already being done in connection with plutonium measurements. Duplicate analyses of plutonium nitrate received from Hanford are required to agree within three parts per thousand. If they do not, one or two additional analyses are run and two or more of the three or four are chosen which satisfy the stated requirement. On this basis, the average difference between duplicates is said to be about 1.6 parts per thousand. However, an additional analysis is run for about

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one fourth of the Hanford batches, implying that about one ninth of all analytical results is rejected. This rejection of data should be carefully reviewed from the statistical point of view, and an effort should be made to determine a more realistic value for the precision of the routine plutonium assays.

5.36 Reports from the counting laboratory include an indication of the reliability of the individual reported values. The reported value is uncorrected for internal absorption. Instead, the absorption effect is added to the "statistical error" to give the reported reliability. These quantities are disparate and should not be combined. Even if knowledge of the amount of internal absorption is believed to be crude, it seems preferable to correct the reported value and to include in the stated reliability only a measure of the uncertainty of the correction (and, of course, the statistical error), rather than the whole value of the correction.

5.37 An opportunity for application of statistical principles occurs in connection with the return to Y-12 of composite samples for isotopic assay. Suitable planning should enable determination not only of the precision of individual assays but also of the presence of other factors, such as changes or trends in standards, adequacy of compositing of the material, adequacy of sampling, and justifiability of rejection of isotopic data at Oak Ridge, though it may be impossible to evaluate these factors separately.

Recommendations

The following recommendations have been discussed with representatives of the Office of Santa Fe Directed Operations and with representatives of the Los Alamos Scientific Laboratory:

a. The contractor should immediately increase his efforts to establish and maintain comprehensive SF material accounting control, including effective coordination of its three basic phases (records, measurements, and reliability).

b. Subordinate recommendations which we believe will be useful in accomplishing the stated major recommendation are that:

(1) The contractor should establish a single accounting control group with full authority over and responsibility for all phases of source and fissionable material accounting control.

(2) Appropriate material measurements which are already being made should be utilized for accounting control to the full extent justified by their reliability.

(3) The records and measurements should be coordinated into a

(4) Additional needed measurements should be made.

In addition, many specific suggestions and recommendations are contained in the body of this report. These should be individually considered by the persons who are assigned responsibility for administering and developing improvements in the system of SF accounting control.

October 15, 1943

Douglas E. George

Horace W. Norton

C. D. W. Thornton

Approved: Walter C. Youngs, Jr.
Walter C. Youngs, Jr.

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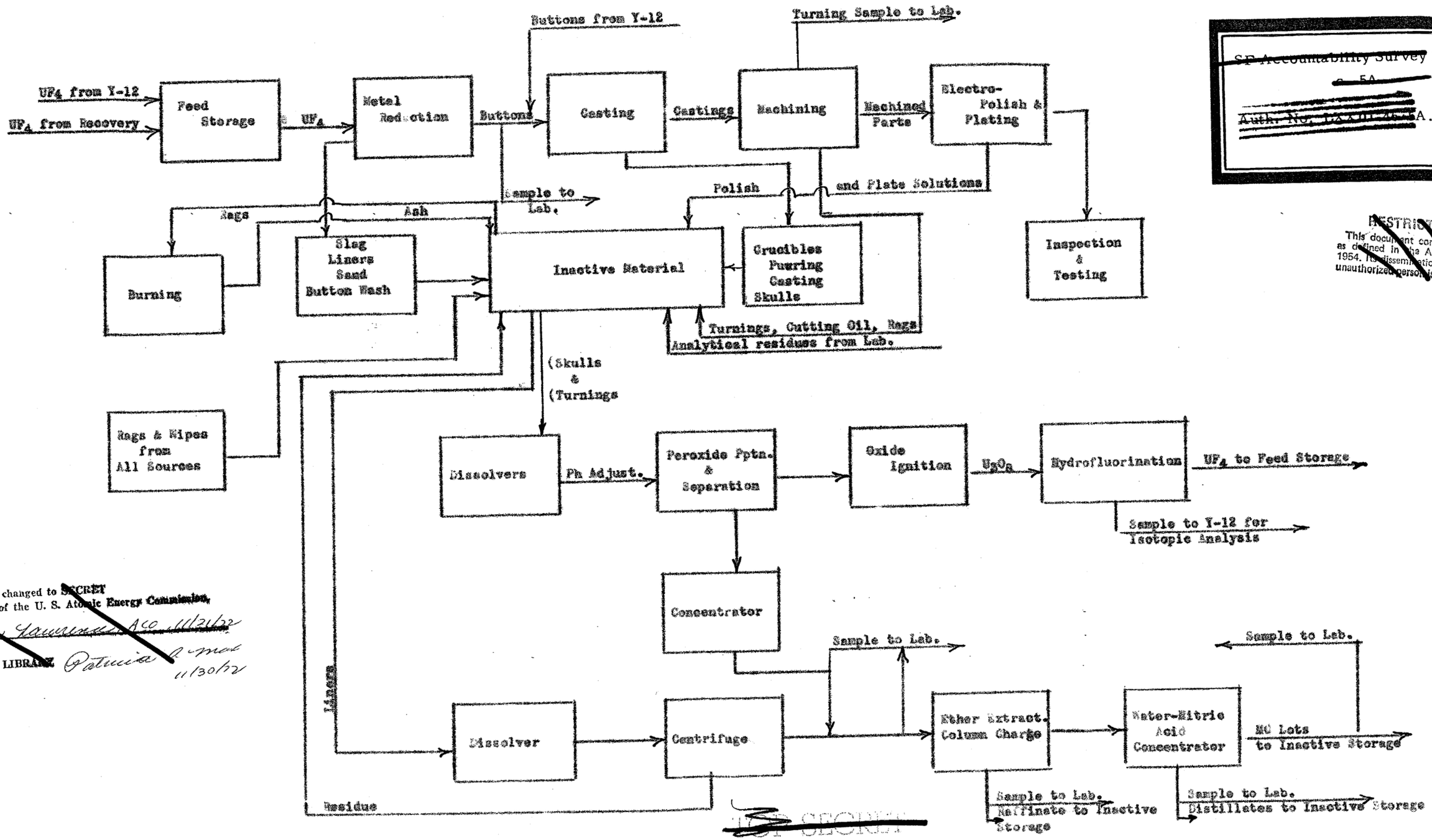
Major Flows of Enriched Uranium



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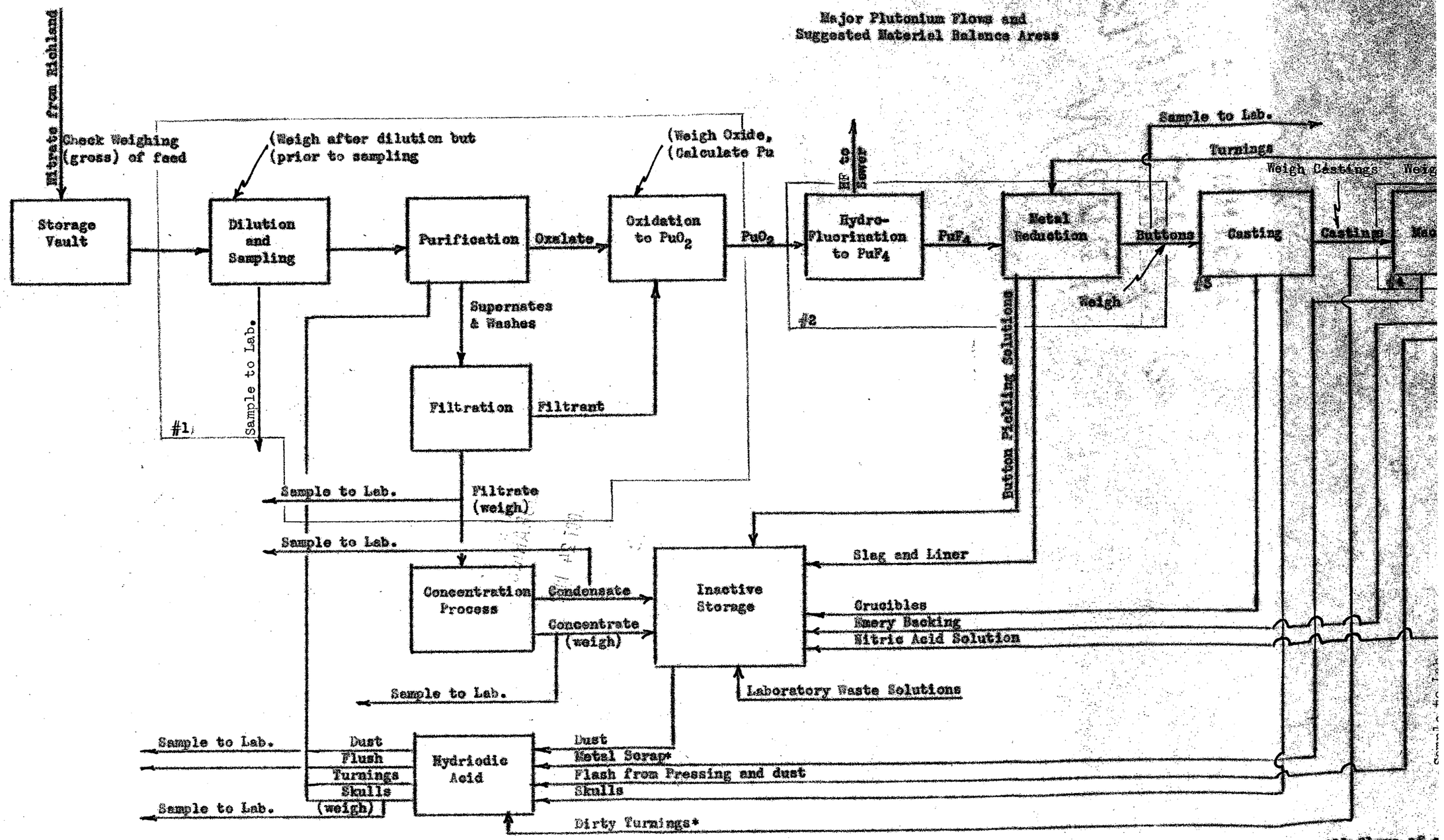
Major Plutonium Flows and Suggested Material Balance Areas



Notes: 1. All flows of s
2. When flows are should be made
3. Numbers on the correspond to

*These flows are identical and duplicate each other.

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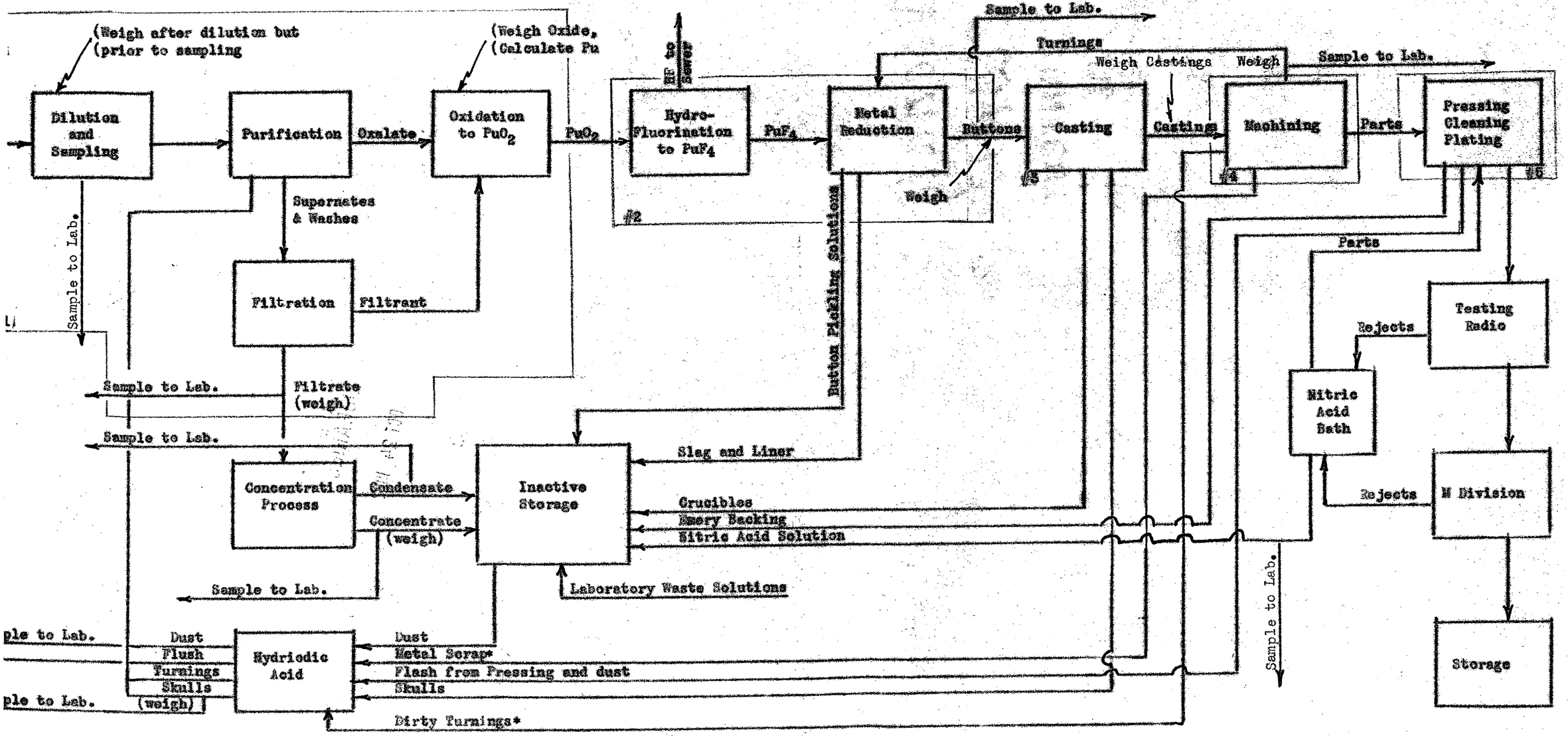
Major Plutonium Flows and Suggested Material Balance Areas



Notes:
1. All flows of samples to laboratory must be accounted for.
2. When flows are interrupted for storage, appropriate entries should be made.
3. Numbers on the above suggested material balance areas correspond to those in text.

*These flows are identical and duplicate each other.

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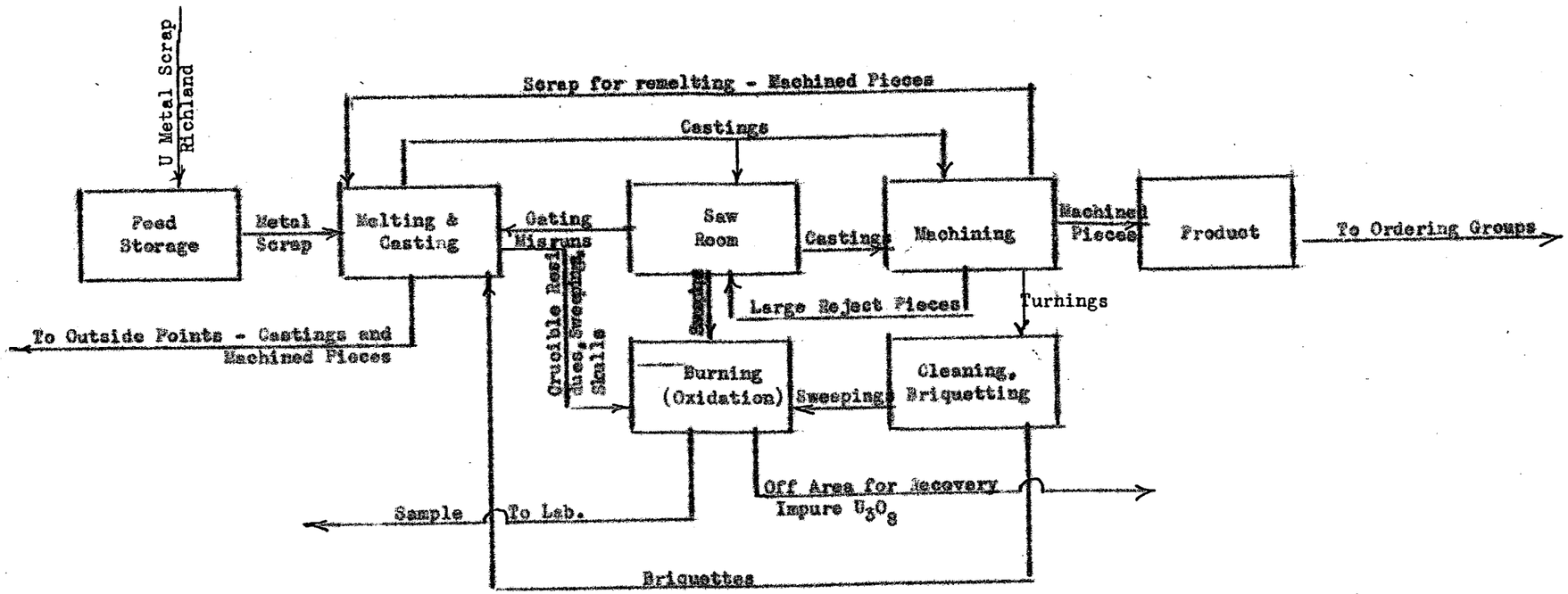
Major Flows of Normal Uranium



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