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AEC AND CONTRACTOR SS MATERIALS MANAGEMENT MEETING

MAY 25–28, 1959

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AEC AND CONTRACTOR

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MAY 25–28, 1959

Issuance date: November 1959

Division of
Nuclear Materials Management
United States Atomic Energy Commission

PREFACE

This report contains papers prepared and presented by Atomic Energy Commission and Contractor Personnel at the Fifth Annual AEC and Contractor SS Materials Management Meeting.

The meeting, sponsored by the Division of Nuclear Materials Management of the Atomic Energy Commission, was held at the Headquarters Auditorium in Germantown, Maryland, May 25-28, 1959, and covered a broad range of problems encountered in AEC and SS Materials Management work.

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AEC URANIUM PROCUREMENT PROGRAM

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Washington, D. C.

Since you are involved in the problems of source and special nuclear materials management, I am assuming that your primary interest in the Division of Raw Materials' program is in our procurement procedures, the contract provisions for delivery, sampling, assaying, payment, etc. AEC source materials accountability begins when the Division of Raw Materials takes title to uranium ores or concentrates, which is the beginning of the production chain. Although our accountability is shortlived, lasting only a few weeks for any given lot of material until it is transferred to the Production Division, this is by no means an adequate measure of the raw materials program. Perhaps the most impressive thing about this program is its recent growth and current size.

Over the next eight-year period we will be purchasing in excess of 200,000 tons of U_3O_8 contained in concentrates having an approximate value of $3.7 billion dollars. It is most important that our techniques of weighing, sampling, and assaying be under continuous review for accuracy and preciseness. An error of as little as one-half of one per cent could mean a dollar difference of $18.5 million.

All our procurement contracts are patterned as nearly as possible after long established commercial practices in the buying and selling of mineral products. For example, long before the atomic program, uranium products had been sold on the basis of U_3O_8. That is why all our production, procurement, assay, and settlement figures are expressed in terms of per cent U_3O_8 content rather than in terms of uranium metal content or mill product.

But before going further into the details of our procurement practices perhaps some information on the background, current status, and future of the raw materials program would be of interest to you.

In 1948 we had an urgent military requirement for uranium. Probably no military program undertaken by this country in peacetime has been considered more important, or been given higher priority, than the atomic weapons program after the breakdown in 1946 of the United Nations Atomic Energy Commission negotiations for the control of those weapons. Events of the succeeding years, particularly in 1949, 1950, and 1951, called for a series of expansions increasing uranium requirements far beyond the capability of any then-known supply.

The AEC's domestic uranium program has been successful. During the past ten years our domestic uranium industry has grown from practically nothing to a position of major importance. The AEC's domestic and foreign uranium purchases in the next three fiscal years will average about 36,000 tons annually, which is in excess of the planned requirements for those years. A uniform delivery rate through 1966, based upon the current estimate of outstanding commitments, would be approximately 27,000 tons a year.

In 1948 nearly all this country's uranium supply came from two small mines, the Eldorado on Great Bear Lake in Arctic Canada, and the Shinkolobwe, Belgian Congo, in the very center of Africa. The production rates of these mines were limited and we could never safely count on ore reserves for more than a few years ahead.

We knew of only three large new sources of uranium-bearing materials, all low-grade, the South African gold ores and our domestic shale and phosphate deposits. In the case of the South African gold tailings, the problem was to develop a process which would recover economically one-quarter to one-half a pound of uranium oxide from a ton of material. Our domestic shales would provide less than one-tenth of a pound of oxide per ton. Our phosphate industry promised only limited production of by-product uranium. By-product recovery from South African gold tailings appeared to be the most promising. However, the AEC undertook extensive research and development for recovery of uranium from all three sources, gold tailings, shale, and phosphate.

There was general concern over our heavy dependence upon Africa for a highly strategic material because of the vulnerability of transportation. In addition to transport from the interior to the coast, the material had to be shipped 7,000 to 10,000 miles across the Atlantic to our East Coast ports. However, we had no choice at that time. Accordingly, arrangements were completed at the end of 1950 for a major South African production program. Except for three small phosphate by-product units, the domestic shale and phosphate programs did not get beyond the research and development stage. Aside from these three low-grade sources, additional large-scale uranium production depended upon new discoveries.

At that time we had almost no domestic uranium production and very few ore reserves. Many of the experts were pessimistic about developing important deposits. No one was thinking in terms of today's production or reserves. In January 1948, developed and partially developed ore reserves on the Colorado Plateau were estimated at approximately 1,000,000 tons containing less than 2,500 tons of recoverable uranium oxide.

In addition to seeking production from every available uranium source, domestic and foreign, programs were established in this country and in Canada to encourage private prospecting and development. Furthermore, the AEC undertook extensive geological investigations and drilling programs to assist domestic development. Special production incentives were established, ore-buying stations were set up, and mine access road programs were sponsored.

The AEC also had to develop new and better processes for milling uranium ores. Its process development program is responsible for the basic processes used today in all our uranium mills. Metallurgical recovery of about 90 per cent has become standard practice. In 1948, the mills recovered only about 70 per cent of the uranium content of the ores.

These metallurgical improvements, together with larger milling operations, have been responsible for lower milling costs and the lower prices now paid by the AEC for concentrate. The average price of domestic concentrates today is \$9.00 per pound of U_3O_8 compared to \$12.50 in 1955. The AEC is reaping benefit from its expenditure of \$26,000,000 for process development.

The first major turning point in the development of uranium reserves on the North American continent came in 1955 with the development of the Blind River field in Canada, discovered in mid-1953. This resulted in Canadian reserves far in excess of our own and provided the basis for a major production effort. Early in 1955 we proposed to limit our concentrate commitment with Canada to properties already under development. This led to the Canadian announcement in August of that year that contracts for mill concentrate would be limited to those companies which could qualify for a mill contract by March 31, 1956. Because of the deep shafts, and extensive preproduction development and construction, full production was not reached until the end of 1958.

We, on the other hand, continued to make contracts for additional domestic concentrate production. Furthermore, in May 1956, the AEC announced a domestic concentrate buying program extending through 1966 to replace the ore buying program begun in 1948, which expires in 1962. This action was based upon a review by the AEC of its projected requirements and purchase contracts, and upon the outlook for additional domestic production that might result from new discoveries. Domestic production was increasing rapidly, but ore reserves were still much too small in relation to this country's current and future requirements.

Figure 1 shows domestic uranium ore reserves by years since January 1948. For the first few years ore reserves increased slowly, but in 1954 the curve started climbing at an accelerating rate. However, at the beginning of 1955, seven years after our domestic program was established, ore reserves still were only 10,000,000 tons, less than two years' supply at today's

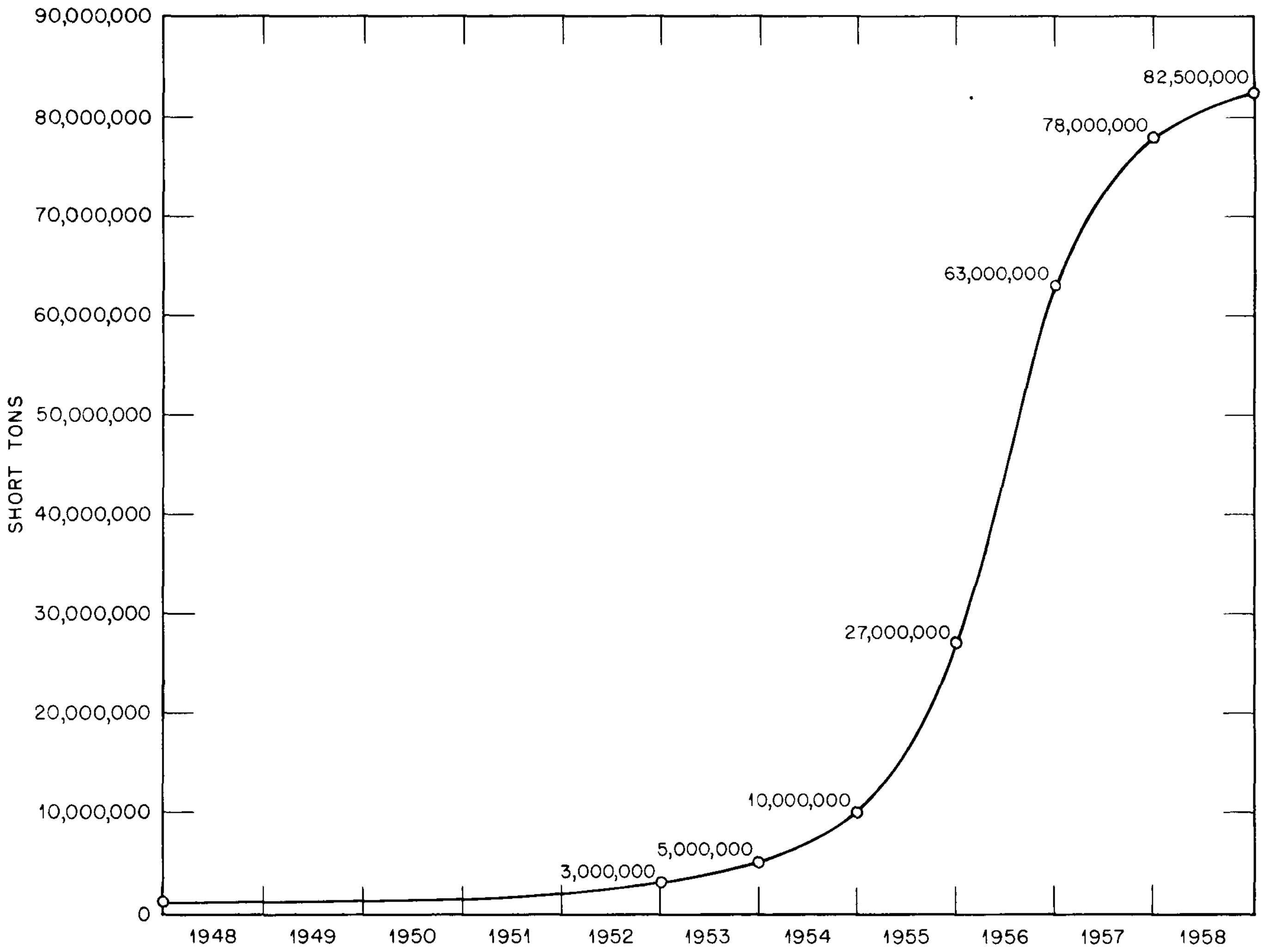

Fig. 1—United States uranium ore reserves (indicated and inferred).

mining rate. At the beginning of 1956 domestic ore reserves were about 30,000,000 tons and they are now estimated at 82,500,000 tons, even though approximately 12,000,000 tons have been mined since 1955.

Figure 2 shows the AEC's uranium purchases by sources, domestic, Canadian, and overseas, beginning with fiscal year 1956. It also shows our current estimates of purchases through 1966 based upon outstanding contracts and commitments. Figures on purchases made prior to fiscal year 1956 are classified, and this is about the only information in the raw materials field, except requirements, that still is classified.

It is evident from these figures that the AEC could not continue to provide a market for production from newly discovered ore reserves. The rapid expansion of production could not be allowed to continue. It would not have been in the interest of the Government or the uranium industry. Consequently, the AEC limited its domestic uranium procurement program by the announcements made on October 28, 1957, and on November 24, 1958, which essentially restricted our purchase commitments to ore already found.

In the current fiscal year domestic production is estimated at 15,150 tons of U_3O_8, more than three times that of 1956. About 50 per cent of current production is from new mills or expanded facilities covered by contracts executed since July 1, 1956. Concentrate production in calendar year 1959 will approximate 18,000 tons of U_3O_8 having a value of more than $300,000,000.

From the beginning of the atomic energy program to July 1, 1955, 83 per cent of the total uranium purchased by the United States came from foreign sources. We had no alternative if the military requirement was to be met. As new sources of uranium were developed on the North American continent, additional purchases were principally from domestic producers and Canada. As I mentioned earlier, in 1955 we limited our commitment with Canada but until the end of 1957 continued to buy all uranium available in the United States.

Between July 1, 1955 and June 30, 1962, 47 per cent of our total uranium purchases will be from domestic sources and 53 per cent from foreign. A large part of the foreign material is Canadian. Most of our Canadian contracts expire March 31, 1962, with only about 2,000 tons to be purchased between March 31, 1962, and March 31, 1963.

Eighty-four per cent of the uranium which the AEC is committed to buy for the period June 30, 1962, through December 31, 1966, will come from domestic sources. The AEC has options for extending its Canadian contracts through 1966 at $8.00 (U.S. currency) per pound of U_3O_8 in a mill concentrate. These options expire March 31, 1961.

With the increased availability of lower cost uranium, prices have come down. The AEC would not consider today, some of the high-cost sources that a few years ago had to be utilized to meet requirements. At one time there was a possibility that we would have to get some of our uranium from low-grade shale and phosphate deposits at a cost of $30 to $50 a pound of U_3O_8.

There has been considerable comment about the prices paid for foreign uranium. AEC contracts for uranium concentrate, domestic and foreign, have been negotiated on the basis of the cost of production, or the estimated cost of production, including amortization, plus a reasonable profit.

An important factor in the higher prices for foreign uranium is the lower-grade ore. On an average, about 9 tons of South African ore must be mined and milled to produce 5 pounds of uranium oxide in concentrate. The same number of pounds of oxide is obtained by mining and milling $2\frac{1}{2}$ tons of average Canadian ore. Only 1 ton of average domestic ore is required to produce 5 pounds of oxide. Whereas, in calendar year 1958, domestic mills processed 5,480,000 tons of ore and recovered 12,500 tons of U_3O_8, South African mills processed 24,200,000 tons of material to produce only 6,146 tons of U_3O_8. A substantial part of this South African production was sold to the United Kingdom. South African uranium is a by-product, or co-product, of gold mining; otherwise uranium costs would be much higher.

When uranium was in short supply it was necessary to make long-range commitments wherever possible and, in addition, to attempt to develop new sources of production to be sure that essential military requirements would be met. Today we are in a position to expand procurement with relatively short lead time. If additional material should be needed, it can be purchased as requirements become firm.

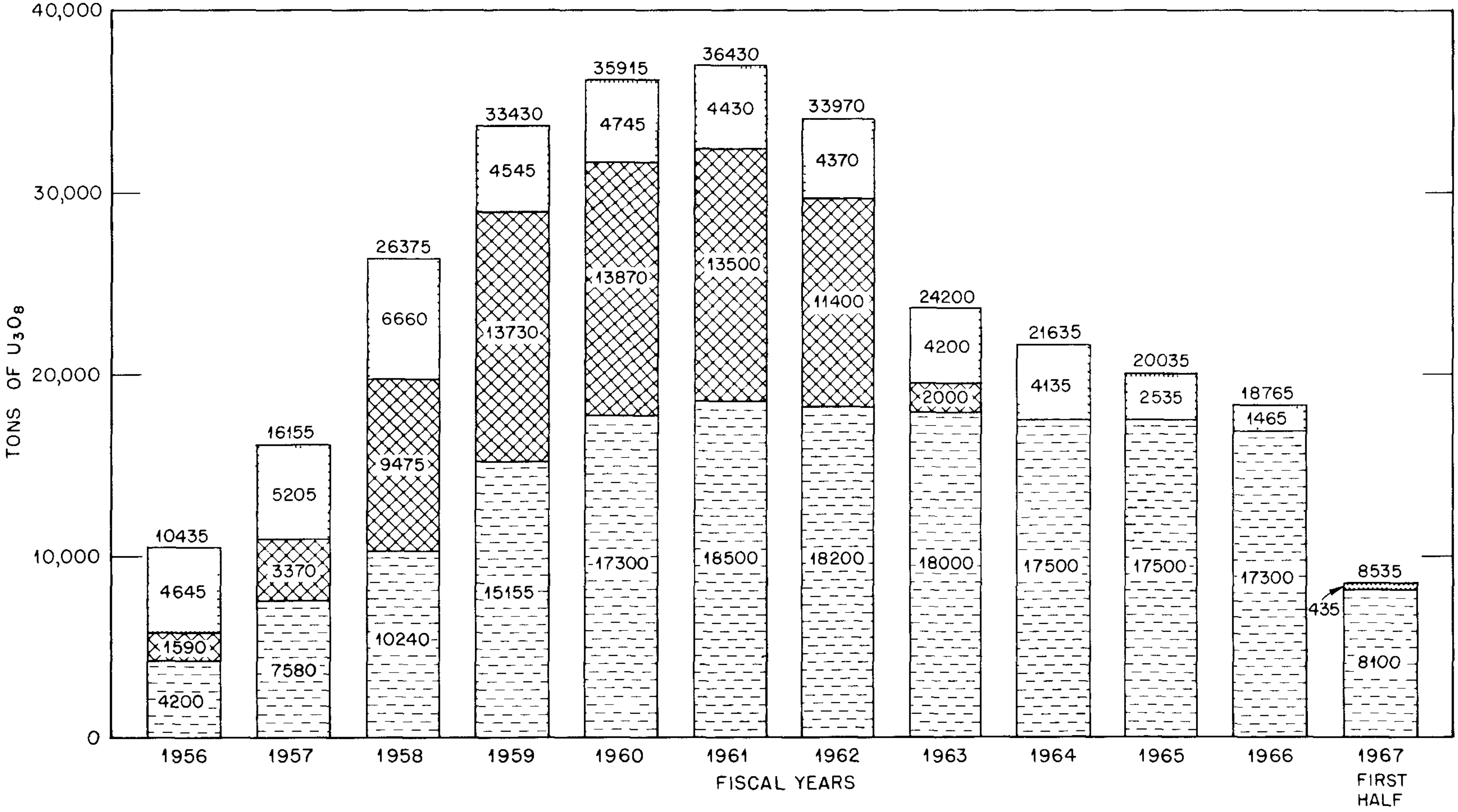

Fig. 2—Uranium deliveries and commitments (☐ overseas, ▩ Canada, ▤ domestic).

The principal developed uranium reserves of the Western Nations today are in the following countries:

	Tons of ore	Tons of U_3O_8
Canada	376,888,000	380,000
South Africa	1,100,000,000	370,000
United States	82,500,000	220,000

France has reported reserves, including potential, of 50,000 to 100,000 tons of uranium.

Although the United States is third on this list, we are in a position to add to our ore reserves more rapidly than almost any other country. Our favorable position is the result of experience and knowledge gained from extensive geological investigations and exploration programs. In this country, we know much more about where and how to search for uranium than we did ten years ago. As yet we have little data upon which to assess the uranium possibilities of much of the world, although we have reason to believe that major deposits will be found in many areas.

Now, I will briefly review our procurement procedures under our major uranium purchase contracts with reference to accountability, sampling, and payment.

Accountability for uranium begins with its initial procurement in a variety of forms and passage of title to the AEC. Domestic uranium is purchased primarily in the form of chemical concentrates, usually impure mixed oxides containing 50 to 90 per cent U_3O_8. Foreign purchases involve both chemical concentrates and metal grade orange oxide (UO_3). Formerly, high-grade ores and mechanical concentrates were also purchased from abroad.

In the case of our overseas procurement, title passes to the Division of Raw Materials F.O.B. vessel, except in South Africa, where it is F.O.B. railway cars at a central calcining plant which I will describe later. Title to Canadian orange oxide passes F.O.B. cars at Port Hope, Ontario, while acceptance of Canadian chemical precipitates does not take place until they reach the AEC feed materials plants. Domestic concentrates acquired by our Grand Junction Operations Office are purchased F.O.B. cars or trucks, Grand Junction. Transfer of accountability from the Division of Raw Materials to the Division of Production for all materials takes place upon receipt at Fernald, Weldon Spring, Paducah, or Oak Ridge.

Following the normal practice of the mining and metal refining industries, contracts for the most part provide for a provisional payment of from 70 to 98 per cent on the basis of the seller's weights and assays. Final payment is made after final weighing, sampling, and assaying in accordance with contract provisions. Contracts provide that the AEC shall weigh and sample each lot of concentrate and that the seller may have a representative present. The AEC and seller each receive a specified number of pulps of each sample and make or cause to be made independent analyses for U_3O_8 content. The results of such analyses are exchanged simultaneously by registered mail. Provision is made for umpire analysis when the assays of the buyer and seller are not within agreed limits.

Originally, the splitting limit used in the assay exchange between the AEC and the producer was 0.4 per cent U_3O_8. This limit is still used for South African concentrates. However, contracts entered into since 1952 provide for an 0.3 per cent splitting limit in most cases. Experience with the Canadian uranium trioxide indicates that an 0.1 per cent splitting limit could be used. This limit is now being used for the uranium trioxide from Belgium.

Contracts set forth specifications for minimum U_3O_8 content and maximum allowable impurities. In the event that the analyses made by the AEC indicate any impurity in excess of specifications, a reduction in price may be required or the lot may be rejected.

The largest source of concentrate, of course, is the Western United States. At this time, there are 23 ore processing mills in operation. With the exception of small amounts of by-product uranium, all domestic concentrate is weighed and sampled at the Grand Junction sampling plant, operated for the AEC by Lucius Pitkin, Inc. Lucius Pitkin personnel tare all empty drums at the mill and stencil the tare weights on the drums. Upon receipt at Grand

Junction, the gross weight of each drum is determined, the contents are auger sampled, and a representative sample is prepared for each lot. Moisture content is determined by drying the sample at 110°C to a constant weight. The dried sample is divided into portions; one split is provided the shipper for his assay, and a second is assayed by Lucius Pitkin for the AEC. These assays are exchanged, and the average taken as the basis for final settlement if they do not differ by more than 0.3 per cent U_3O_8. If they differ by more than this amount, an umpire analysis on a third sample split is performed by the National Bureau of Standards and the middle of the three assays is used for settlement.

To date, the Western mills have not generally exercised their right to have a representative present for official weighing, sampling, and sample preparation. Company representatives have visited and examined the Grand Junction sampling plant and its procedures, however, and have indicated satisfaction with the operation.

Canada is the largest source of foreign uranium, supplying the AEC about 14,000 tons U_3O_8 per year. Approximately 3,800 tons of this material is in the form of metal grade uranium-trioxide produced at the Port Hope, Ontario, refinery of Eldorado Mining and Refining, Limited, a government-owned corporation. The remainder is in the form of mill concentrates containing 50 to 80 per cent U_3O_8. These are shipped to the Fernald or Weldon Spring feed materials plants where weighing and sampling is performed. Gross weights are obtained and the stencilled tare weights on the Canadian drums are checked on a statistical basis. The drums are auger sampled and assay samples are prepared from a bulk sample of each lot and distributed. Ledoux and Company represents the Canadian producers at the weighing and sampling.

Assays are made on an "as is" basis; that is, the sample is analyzed without drying and the impurity contents expressed as per cent of the uranium content. This eliminates a moisture loss determination which entails certain precautions under conditions of widely fluctuating ambient humidity. The assay results are exchanged with Canadian producers, and the National Bureau of Standards is the umpire laboratory.

Weighing and sampling of the Canadian uranium trioxide is done at the Port Hope refinery. The Division of Raw Materials has a representative at Port Hope who spot checks the gross and tare weights of the drums, checks the scales, and observes the sampling and sample preparation. The uranium trioxide product is continuously sampled by a horizontal screw auger from the stream falling into the containers. The sampling system is protected from the atmosphere and the assay sample is prepared on an "as is" basis after proper blending. The assay samples are sealed in bottles and sent with the shipment to either Paducah or Oak Ridge. Exchange of assays and provisions for umpire analysis are the same as for the Canadian mill concentrates.

South Africa represents the third largest source of concentrates. Seventeen plants in the Union of South Africa produce chemical concentrates by an ion exchange process. These are delivered in the form of thickened slurries in tank trucks to a central calcining plant, Calcined Products (Pty.), Ltd. Here the material is dried, sampled, assayed, and packed for shipment as designated by the Combined Development Agency, a joint organization of the U.S. and U.K. governments for the procurement of uranium. The incoming slurry is filtered and the filter cake is extruded, dried, and calcined at 500°C. Each ten-ton lot is continuously sampled during the packaging operation to provide a bulk sample of about one per cent. A representative of the Combined Development Agency is permanently stationed at the plant to observe and check on the official weighing and sampling.

A portion split from the bulk sample is calcined at 900°C to obtain a stable material for analysis and the weight loss on calcining is reported and used for calculation of the U_3O_8 content. Assay samples are prepared and shipped via air express to the Division of Raw Materials, Washington, and then forwarded on to the Fernald, Ohio, plant where the official U.S. AEC assays are made. Assays are exchanged with Calcined Products; if umpire assays are required, the National Bureau of Standards and the South African Government Metallurgical Laboratory are the alternate umpires.

We also purchase smaller quantities of uranium from the Belgian Congo, Australia, and Portugal. In the past the Belgian Congo has shipped both mechanical concentrates and mill precipitates of similar grade to domestic concentrates. However, remaining deliveries under the contract will be in the form of uranium trioxide. This material will be shipped to Oak Ridge for weighing and sampling. The Belgians have a representative present at these operations.

It would be impossible in the time allotted to discuss all of the details of the development of our uranium procurement program and of the general purchase procedures described. However, I am pleased to have had the opportunity to give you this brief summary.

USE OF AUTOMATIC DATA-PROCESSING EQUIPMENT IN AN ANALYTICAL QUALITY CONTROL PROGRAM AT THE SAVANNAH RIVER PLANT

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Introduction

Since the accuracy and precision of SS materials accountability are dependent on the quality of chemical and isotopic analyses, it is necessary to have rigid control over the reproducibility and accuracy of the analytical values.

At the Savannah River Plant we have had an analytical quality control program since Plant startup. Prior to 1956, bias and precision were determined by individual Plant laboratories through analyses of standard or synthetic samples. Types of control charts, methods of determining control limits, and types of statistical reports varied for different laboratories and for different types of analyses.

Due to the need for uniform, reliable statistics, the current Plantwide quality control procedure was established in the spring of 1956. This procedure was designed to provide continuing control over both the accuracy and the precision of analytical values reported by the various laboratories, while at the same time freeing laboratory personnel from time expended in statistical calculations and interpretation.

Specific requirements for the procedure were that it provide:

- Prompt and accurate reports, on a monthly and quarterly basis, of the bias and precision of all analyses important to process control and accountability, with an indication of the significance of changes in bias and precision.

- Control charts and control limits which would enable laboratory personnel to maintain immediate and continuing control and interpretation of analytical results.
- A simple routine for recording and plotting the minimum number of determinations necessary for reliable statistics and continuing control.

A preliminary study was made of the precision of the statistics obtainable from varying numbers of determinations, with special attention given to accountability samples. This study, together with a review of the sampling points and chemical analyses essential to accountability and process control, indicated a larger quality control program and more calculations than could be handled manually by available statistical personnel. Therefore, the decision was made to utilize available data-processing equipment.

Previous statistical studies and quality control data were used to obtain estimates of the number of significant digits to be recorded and reported. Forms for recording and plotting data were then designed, taking into consideration the number of items to be included, the number of digits in each item, and the types of statistical reports which could be prepared from the data.

Since all levels of concentrations and impurities occuring in production samples are not reproducible in synthetic samples, the decision was made to include independent duplicate analyses of routine production samples in the quality control program to determine precision. Synthetic samples were included for determination of bias and for checking precision calculations.

In any statistical program, the data to be collected and the statistics to be calculated are determined not by the methods used to collect the data or perform the calculations, but rather by the uses to be made of the data and the types of final reports to be prepared. Use of data-processing equipment ensures rapid and accurate calculations and permits examination of data in a greater variety of ways than would be feasible with manual calculations, but the program is essentially the same regardless of the methods used to calculate statistics. Today I shall describe the procedure instituted at the Savannah River Plant in 1956, emphasizing the role of statistical calculations and data-processing equipment.

Data-Processing Equipment

When the procedure was designed, available machines consisted of conventional IBM accounting equipment and electrical desk calculators. In October 1958 an IBM 610 electronic computer became available, and in January 1959 an IBM 650 was acquired. Calculations previously performed on the electrical desk calculators were transferred to these machines. Final reports are now produced on the IBM 650. These transitions were made easily, even though the procedure was not originally designed for use with the 610 or 650. Our experience indicates that the ease of transition to automatic computers is directly related to the care with which the procedure was originally planned.

Since conventional data-processing equipment is available to most of you, I shall describe the initial procedure, pointing out operations transferred from statistical personnel when the automatic computers became available.

IBM equipment available in the spring of 1956 included the following electrical machines:

> Card Punching Machines to transcribe data from source records into IBM cards in punched hole form and to verify punched data.
>
> Sorters to arrange punched cards in sequence according to any classification punched in the cards.
>
> 604 Calculating Punch to perform and punch simple calculations into cards.
>
> 407 Punched Card Accounting Machines to prepare and print reports and records after the cards have been arranged in the required sequence, and to add or subtract, printing combinations of totals as planned.

In any quality control program a certain amount of consistency in recording data is desirable. When data are to be punched into cards, such consistency is essential. Therefore, standard forms were designed for use throughout the Plant in recording analytical results. To simplify punching of data, card column numbers were printed on the forms. For security reasons and to simplify sorting of data, numerical codes were assigned to each type of analysis and each sample point. These codes were assigned by the statistical group, and are used to control IBM reports.

Although the IBM procedure was specifically designed for quality control data, it has often been used for statistical analysis of data outside the routine quality control program. This flexibility was achieved by designing a generalized program controlled by assigned numerical codes.

I shall now present examples and explanations of the forms, IBM card format, IBM reports, statistical worksheets, and final statistical reports as initially designed for the quality control program.

Analyses of Synthetic Samples

As previously stated, standards or synthetic samples are analyzed to determine both accuracy and precision. The forms used to plot and record data are shown in exhibits 1A and 1B. In cases where analyses are performed in duplicate and averaged to obtain the value reported by the laboratory, both the reported value and the range in duplicates are entered on the forms.

Twenty initial analyses per month are performed at equal chronological intervals. When possible, the same standard sample is used for the three-month period covered by the control chart (exhibit 1A). Control limits are based on the previous quarter's performance, and remain in effect for the entire current quarter unless monthly summaries indicate a significant change in bias or precision. When an analysis is outside control limits, it is repeated once and both values are recorded for quality control purposes. If the second analysis is also out of control, supervision is notified, and an attempt is made to ascertain the cause. The same sample number, shift, and date apply to the repeated analysis, when the initial analysis is out of control.

Once a month, the completed data form (exhibit 1B) is submitted to the statistical group, where the identification of the analysis and method, sampling point, and unit of measurement are coded and entered in the block in the upper right corner of the form entitled "IBM Use Only." The form is edited, and a one-digit code is assigned to each analysis indicating whether the analysis is in or out of control and whether it is an initial or repeated determination.

The form is then submitted to the machine accounting group. Exhibit 2A illustrates the IBM card format. Items in columns 1 through 35 are punched directly from the form; items in the remaining columns are punched by the 604 calculator and consist of the difference in the reported value and the standard value (with sign), the square of the difference, the square of the reported value, the square of the range, and the range in duplicates expressed as a percent of the reported value. The cards are then sorted, and three types of reports are issued monthly and quarterly for each analysis and method:

- Frequency distributions of differences in the reported value and standard value, for selected groupings of data. (A typical report of this type is shown in exhibit 2B.)
- A listing of all information for reference purposes.
- Selected sums and sums of squares to be used to calculate final statistics.

Prior to the acquisition of the 650, final statistical reports were prepared by the statistical group using the IBM reports. Examples of worksheets used by statistical personnel are shown in exhibits 3A and 3B. "Initial" laboratory determinations are used for the first set of statistical calculations. Extreme deviations are deleted when Chi Square calculations indicate that the statistics are invalid for the majority of the data. Repeated analyses are substituted for deleted initial analyses, and the process is continued until statistics are obtained representing the best estimate of the bias and precision of values reported by the laboratory.

The formulas used in the calculations are printed on the worksheets. Ungrouped data formulas are used to calculate mean and variance. In the selection of formulas, consideration was given to the machines to be used in computations.

Until the 610 was acquired, the calculations were performed by clerks who had been trained to use desk calculators. The relationship of the frequency distributions and statistics enabled one statistician to review the calculated statistics with a minimum of effort and with confidence in the validity of the calculations. Examples of the types of final reports prepared by the statistical group are shown in exhibits 4A and 4B.

Duplicate Analyses of Routine Production Samples

The procedure for duplicate analyses of routine production samples is similar to the procedure for synthetic samples. Twenty routine production samples are analyzed independently by two different shifts each month, to determine precision of analytical determinations. The initial analytical value is reported in a routine manner. The analytical value obtained by the second shift is compared with the value reported by the first shift, by plotting the two values as one point on a control chart (exhibit 5A). When the plotted point falls outside the control limits, the duplicate analysis is repeated once. If the point plotted using the repeated value is also outside of control limits, an attempt is made to determine the reason.

This phase of the over-all quality control program differs from the synthetic-sample phase in that total variance in reported values is governed largely by variations in the production process, and the basic aim, from a quality control standpoint, is to control that portion of the variance which is due to error in the analytical determinations. Since there is a significant positive correlation in analytical results obtained by the same person on the same sample, and since in some instances only one person on a shift performs a particular type of analysis, the procedure requires that a different shift perform the duplicate determination. To assure representative samples, shift rotation and chronological aspects are considered in predetermining production samples to be included in the program.

For each sample analyzed in duplicate, there is one degree of freedom in calculating variance; therefore, for 20 samples or 40 determinations, there would be 20 degrees of freedom. It would appear that approximately twice as many analyses are necessary to calculate precision as are necessary with synthetic samples; but since half of these are routine analyses of production samples, the number of analyses for quality control purposes is actually about the same.

As opposed to analyses of a synthetic sample, we are not dealing with the subtraction of a constant from variables, but with differences between paired variables.

Let X_i and Y_i represent duplicate values obtained on the i^{th} sample, and let $\Delta_i = |Y_i - X_i|$; then:

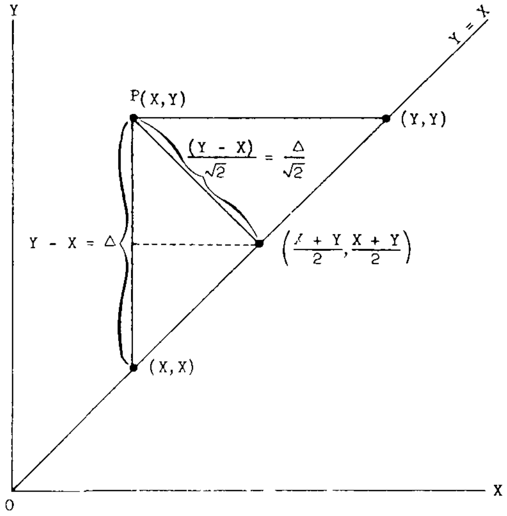
$$\left(X_i - \frac{X_i + Y_i}{2}\right)^2 + \left(Y_i - \frac{X_i + Y_i}{2}\right)^2 = \left(\frac{X_i - Y_i}{2}\right)^2 + \left(\frac{Y_i - X_i}{2}\right)^2 = \frac{\Delta_i^2}{2}$$

For one pair, $\frac{\Delta}{\sqrt{2}}$ is an estimate of the standard deviation, with one degree of freedom. For N pairs, there are N degrees of freedom, so:

$$\sigma^2 = \frac{\sum_{i=1}^{N}\Delta_i^2}{2N} \qquad \text{and} \qquad \sigma = \frac{1}{\sqrt{2}}\sqrt{\frac{\sum_{i=1}^{N}\Delta_i^2}{N}}$$

If the 95% confidence interval ($\pm 1.96\sigma$) is used as control limits, the maximum difference permitted in a pair of analytical determinations is $(\sqrt{2})(1.96\sigma)$. This precision is used as the basis for the control limits shown on the control chart (exhibit 5A).

Using the same scale for the reported value (X) and the duplicate analysis (Y), any pair of values can be plotted as a point $P(X,Y)$. A perpendicular from P to the line $Y = X$, **intersects** the line at the point $\left(\frac{X+Y}{2}, \frac{X+Y}{2}\right)$, which represents the mean of the duplicate analyses. The length of this perpendicular distance is $\frac{Y - X}{\sqrt{2}} = \frac{\Delta}{\sqrt{2}}$.



When the precision is expressed as percent, control limits for control charts can be drawn by multiplying points on the line Y = X by the precision. These perpendicular distances from the points on the line Y = X determine points for the control limits. (The same control limits can be established, of course, by dividing the amount determined by the product of the values and the precision by the square root of two, and measuring horizontally and then vertically from the points on the line Y = X.)

When the distribution of analytical values reported by the laboratory is significantly skewed, the precision expressed as a percent of the average reported value can be misleading. Therefore, differences of each pair are calculated as a percent of the mean of the duplicates, and distributions of the percents are analyzed statistically, as well as the distributions of arithmetic differences in duplicates.

The form used to record duplicate analyses of routine production samples is shown in exhibit 5B. The data recorded on the form are similar to those recorded for synthetic samples (exhibit 1B), and the coding procedure used by the statistical group is similar to that used for coding the synthetic-sample data.

Forms are submitted to the machine accounting group, where cards are keypunched, verified, calculated, sorted, and tabulated. Again, three types of IBM reports are issued monthly and quarterly for each analysis and method:

- Frequency distributions, for selected groupings of data, of: (a) arithmetic differences in duplicates, (b) arithmetic means of duplicates, and (c) differences expressed as percents of means.
- A listing of all information for reference purposes.
- Selected sums and sums of squares for use in statistical calculations.

These IBM reports are used by statistical personnel to prepare final reports. Precision calculations can be made using either the sums of squares of the percent differences, or the sums of squares of the arithmetic differences. Because of the variance in the production process, the calculations are usually based on the sums of squares of the percent differences; and the median reported value, rather than the arithmetic mean, is reported. (The distributions of reported values are helpful in establishing the concentration of comparable synthetic samples.) Chi Square analysis is applied to the frequency distribution of initial percent differences, and extreme values are deleted and replaced by repeated values until reliable precision figures are calculated. An example of the type of final report prepared by the statistical group is shown in exhibit 6.

Utilization of Automatic Computers

In conclusion, I shall review briefly the operations transferred to automatic computers.

The IBM 610 computer was acquired in October 1958. At that time all calculations previously handled by statistical personnel were transferred to the 610. Programing and operation of the 610 are simple; calculations are performed automatically and are printed on a high-speed typewriter.

However, the original procedure was retained intact, and only final statistical calculations performed by the statistical group were transferred to the 610.

The IBM 650 was acquired in January of this year. In March, all calculations, decisions, and reports previously made by statistical personnel were transferred to this computer. Forms for recording and plotting data remain the same, but coding of initial and repeated analyses is simplified for the statistical group, in that it is not necessary to indicate whether the analysis is in or out of control.

Since the addition of the 650, generalized programs have been prepared, to handle analysis of variance and more involved statistical calculations; thus there is not now the need to use the IBM quality control procedure to process other data. However, as previously mentioned, the initial IBM procedure has been used for numerous other statistical studies. For accountability purposes, studies were made of error or variance due to sampling, temperature, instrumentation, production operator error, and laboratory determinations at accountability sampling points. More than 200 different types of analyses or special data have been processed using this procedure, and for nearly three years it has fulfilled a need at the Savannah River Plant by enabling a small statistical group to routinely handle, in short periods of time, large volumes of quality control data for all Plant laboratories.

List of Exhibits

EXHIBIT 1A

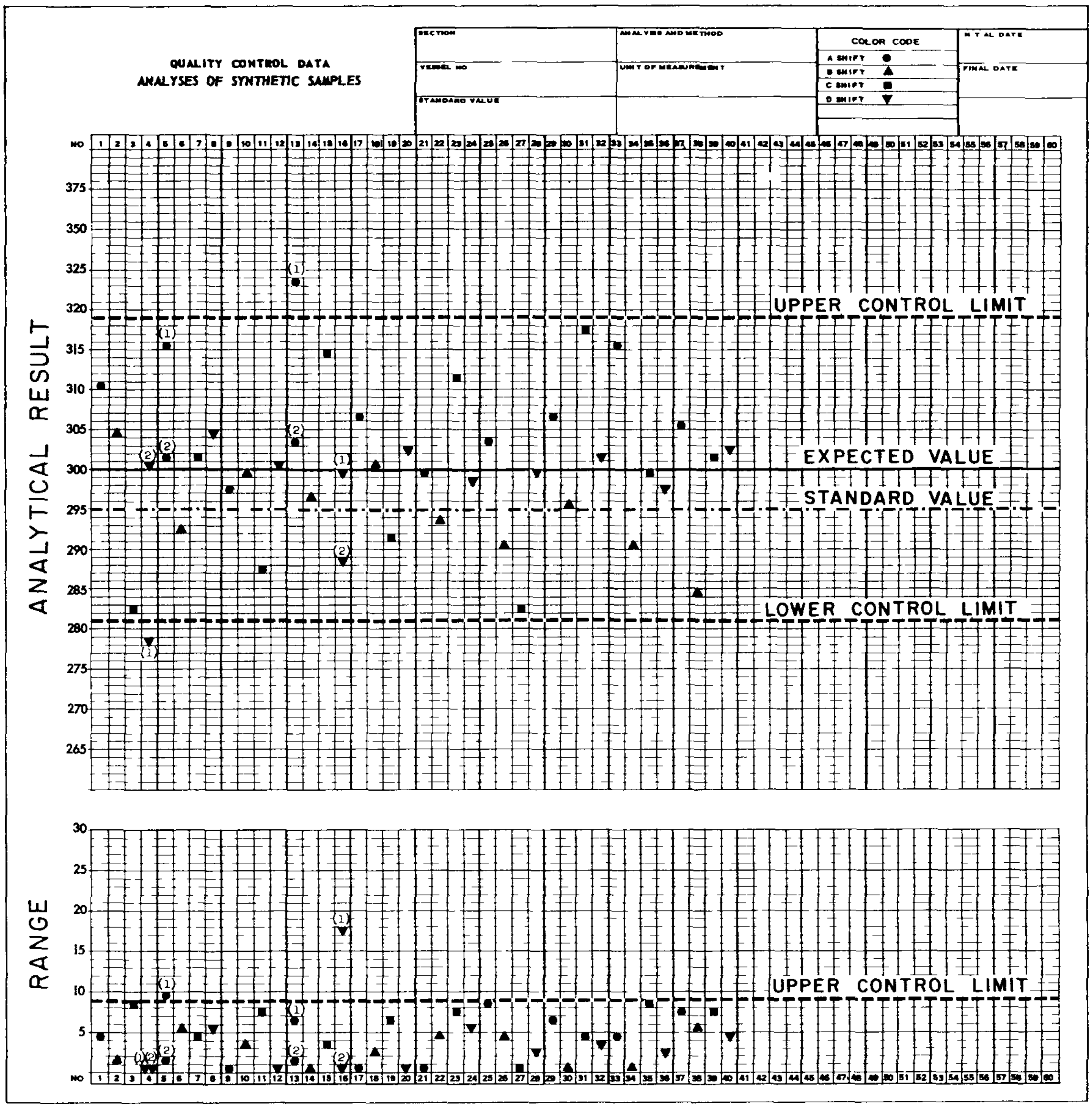

QUALITY CONTROL DATA
ANALYSES OF SYNTHETIC SAMPLES
SECTION
VESSEL NO
STANDARD VALUE
ANALYSIS AND METHOD
UNIT OF MEASUREMENT
COLOR CODE
A SHIFT ●
B SHIFT ▲
C SHIFT ■
D SHIFT ▼
INITIAL DATE
FINAL DATE
NO
ANALYTICAL RESULT
375
350
325
320
315
310
305
300
295
290
285
280
275
270
265
UPPER CONTROL LIMIT
EXPECTED VALUE
STANDARD VALUE
LOWER CONTROL LIMIT
(1)
(2)
RANGE
30
25
20
15
10
5
UPPER CONTROL LIMIT

EXHIBIT 1B

QUALITY CONTROL DATA
ANALYSES OF SYNTHETIC SAMPLES

INITIAL DATE ______ __

SECTION

ANALYSIS AND METHOD

UNIT OF MEASUREMENT

VESSEL NUMBER

STANDARD VALUE

IBM USE ONLY		
FIELD	IBM COL NO	CODE
A	1 – 2	2
B	3 – 4	
C	5 – 6	
D	7 – 9	
E	10	1
F	11 – 15	

SHIFT	NO
A	1
B	2
C	3
D	4
DAYS	5

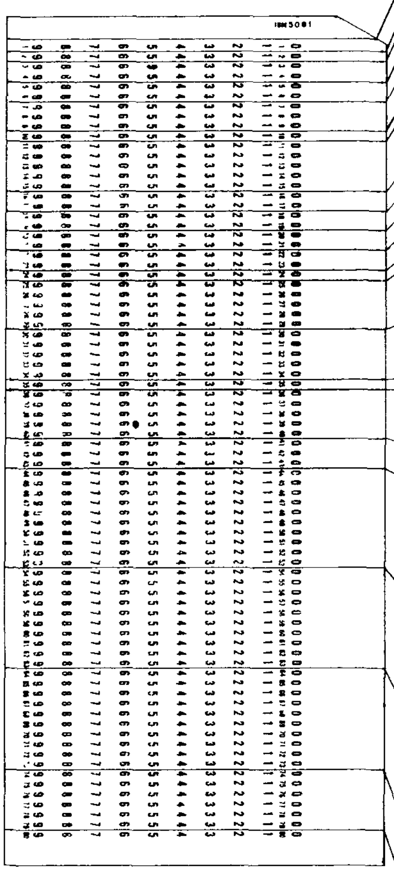
FIELD	G	H			I	J	K	L
IBM COL NO	16 – 17	18 – 23			24	25 – 29	30 – 34	35
LAB USE ONLY SAMPLE NO	IDENTIFICATION NUMBER	DATE MO.	DAY	YEAR	SHIFT NO	REPORTED VALUE	RANGE IN DUPLICATES	IBM USE ONLY

EXHIBIT 2A

QUALITY CONTROL DATA - ANALYSES OF SYNTHETIC SAMPLES

IBM Card Format

(▶ indicates decimal point.)



1	Type of Data	KEYPUNCHED FROM DATA FORM (EXHIBIT 1B)
2	Section	
3 4	Analysis and Method	
5 6	Unit of Measure	
7 8 9	Sample Identification	
10	IBM Card Code	
11 12 13 ▶ 14 15	Standard Value	
16 17	Sample Number	
18 19	Month — Date	
20 21	Day — Date	
22 23	Year — Date	
24	Shift	
25 26 27 ▶ 28 29	Reported Value	
30 31 32 ▶ 33 34	Range in Duplicates	
35	In or Out of Control	
36 37 38 ▶ 39 40	Δ = Reported Value - Std Value	CALCULATED AND PUNCHED BY IBM 604 CALCULATOR
41 42 ▶ 43	% Difference in Duplicates	
44 45 46 47 48 49 ▶ 50 51 52 53	(Reported Value)^2	
54 55 56 57 58 59 ▶ 60 61 62 63	(Range)^2	
64 65 66 67 68 69 ▶ 70 71 72 73	Δ^2 = (Reported Value - Standard Value)^2	
74 75 76 77 78 79		
80	Month Code	

EXHIBIT 2B

QUALITY CONTROL DATA - ANALYSES OF SYNTHETIC SAMPLES

Dates of Analyses: ____________________

SECTION ____________________
ANALYSIS AND METHOD ____________________
SAMPLE IDENTIFICATION ____________________
UNIT OF MEASUREMENT ____________________

Identification (Numerical Code)	Reported Value - Standard Value	Number of Analyses						
		In Control	Out of Control					
		Initial and Repeat	Mean In, Range Out		Mean Out, Range In		Mean Out, Range Out	
	Δ		Initial	Repeat	Initial	Repeat	Initial	Repeat
1 41 211 0	.77-					1		
	.68-				1			
	.67-				1			
	.60-				2			
	.58-				1	1		
	.52-				1	2		
	.50-					1		
	.42-				1	1		
	.40-					1		
	.37-				1	1		
	.36-				1	2		
Lower Control Limit, Previous Quarter	.35-				2	1		
	.34-	3						
	.33-	2						
	.32-	2						
	.31-	1						
	.30-	1						
	.26-	4						
	.25-	2						
	.24-	1						
	.22-	3						
	.21-	1						
	.20-	2						
	.18-	1						
	.16-	3						
	.14-	1						
	.13-	2						
	.12-	2						
	.08-	1						
	.05-	1						
	.02-	3						
	.01-	1						
	.05	1						
	.08	1						
	.10	1						
	.12	1						
	.13	3						
	.14	1						
	.17	2						
Upper Control Limit, Previous Quarter	.22	1						
	.25					1		
	.32				1			
	.34				1			
	.87				1			
	Total	48*	*	*	14*	12*	*	*

EXHIBIT 3A

QUALITY CONTROL DATA - ANALYSES OF SYNTHETIC SAMPLES

Statistical Worksheet for Calculating Bias and Precision

for Quarter Ending ____________

SECTION ______________________
ANALYSIS AND METHOD ______________________
SAMPLE IDENTIFICATION ______________________
UNIT OF MEASUREMENT ______________________

N ______________
N - 1 ______________
N(N - 1) ______________
$\sqrt{N}$ ______________

	Total Number	Number Included in Calculations
Total Initial Analyses		
Within Control Limits		
Out of Control Limits		
Repeated Analyses		
Within Control Limits		
Out of Control Limits		
TOTAL		

	Sum	Sum of Squares	Arithmetic Mean	Variance	Standard Deviation	Precision (95% C. I.)	
				$\sigma^2 =$		Arithmetic Amount	% of R. V.
	ΣX	ΣX^2	$\bar{X} = \frac{\Sigma X}{N}$	$\frac{N\Sigma X^2 - (\Sigma X)^2}{(N)(N - 1)}$	$\sigma = \sqrt{\sigma^2}$	1.96σ	$\frac{(1.96\sigma)(100)}{R.V.}$
Standard Value							
Reported Value							
Δ = R.V. - Std. V.							
Range in Duplicates							

Expected Bias = $\bar{\Delta}$ = $\overline{\text{Reported Value}} - \overline{\text{Standard Value}}$ = ______

Percent of $\overline{\text{Reported Value}}$ = $\frac{(\bar{\Delta})(100)}{\overline{R.V}}$ = ______%

SIGNIFICANCE OF BIAS:

$t = \frac{(\bar{\Delta})(\sqrt{N})}{\sigma} =$ ______

Degrees of Freedom = N - 1 = ______

Probability that bias equals zero:

__________ $\leq P \leq$ __________

TESTS FOR SIGNIFICANCE OF DIFFERENCES IN PRECISION AND BIAS CALCULATED FOR CURRENT QUARTER AND PREVIOUS QUARTER

	i	N_i	$N_i - 1$	σ_i^2 = Variance	$\bar{\Delta}_i$ = Bias
Previous Quarter	1				
Current Quarter	2				

PRECISION

RATIO OF VARIANCES: F TEST

$F_{(N_1 - 1,\, N_2 - 1)} = \frac{(\qquad)}{(\qquad)} =$ __________

Within 95% Confidence Interval, precision for current quarter is

significantly better than	☐	(Check One)
significantly worse than	☐	
not significantly different from	☐	

precision calculated previous quarter.

BIAS

$$t = \frac{\bar{\Delta}_2 - \bar{\Delta}_1}{\sqrt{\left[\frac{(N_1 - 1)\sigma_1^2 + (N_2 - 1)\sigma_2^2}{(N_1 - 1) + (N_2 - 1)}\right]\left[\frac{1}{N_1} + \frac{1}{N_2}\right]}}$$

$$t = \frac{(\qquad) - (\qquad)}{\sqrt{(\qquad)(\qquad)}} = \frac{(\qquad)}{(\qquad)}$$

t = __________ m = $N_1 + N_2 - 2$ = ____

Probability that bias is not significantly different from previous quarter:

__________ $\leq P \leq$ __________

FOR CONTROL CHARTS (Add to Standard Value):

BIAS = ______ (Expected Average)
BIAS - 1.96σ = ______ (Lower Control Limit)
BIAS + 1.96σ = ______ (Upper Control Limit)

Range in Duplicates:

Average Range ______
Lower Control Limit 0
Avg Range + $1.96\sigma_{Range}$ = ______ (Upper Control Limit)

REMARKS: ____

____

Calculations by __________
Date __________

EXHIBIT 3B

QUALITY CONTROL DATA - ANALYSES OF SYNTHETIC SAMPLES

Statistical Worksheet for Testing Normality of Distribution of Deviations of Reported Value from Standard Value

for Quarter Ending ____________

SECTION ______________________
ANALYSIS AND METHOD ______________________
SAMPLE IDENTIFICATION ______________________
UNIT OF MEASUREMENT ______________________

Reported Value	Range in Duplicates	Number of Analyses			Number of Analyses Used to Test Distribution		
		Total	Initial	Repeats	Total	Initial	Repeats
In Control	In Control	____	____	____	____	____	____
Out of Control	Out of Control	____	____	____	____	____	____
In Control	Out of Control	____	____	____	____	____	____
Out of Control	In Control	____	____	____	____	____	____
	TOTAL ANALYSES	____	____	____	____	____	____

EXPECTED BIAS = $\overline{\text{REPORTED VALUE}} - \overline{\text{STANDARD VALUE}} = \bar{\Delta}$ = ____________

STANDARD DEVIATION OF REPORTED VALUE = σ = ____________

①	②	③	④	⑤	⑥	⑦	⑧
Expected Distribution			Range in Δ		N = ____	Actual Frequency	Chi Square Values
Range in Δ		Frequency	Minimum	Maximum	Expected Frequency (Use ③)		$\frac{(\text{⑦} - \text{⑥})^2}{\text{⑥}}$
Add to Δ for:							
Minimum	Maximum		$\bar{\Delta}$ + ①	$\bar{\Delta}$ + ②			
Less than or equal to -1.65σ		.05N					
-1.64σ	-1.28σ	.05N					
-1.27σ	$-.52\sigma$	.20N					
$-.51\sigma$	$.00\sigma$	.20N					
$.01\sigma$	$.52\sigma$	.20N					
$.52\sigma$	1.27σ	.20N					
1.28σ	1.64σ	.05N					
1.65σ or more		.05N					

Degrees of Freedom = ______ Chi Square = ______

Probability that distribution is normal for mean and standard deviation tested: __________

ASSUMPTION:

$\bar{\Delta}$ = __________ σ = __________

Condition	Number of Analyses	Probability		
$\bar{\Delta} - 1.96\sigma > \Delta > \bar{\Delta} + 1.96\sigma$	________	________	$\leq P \leq$	________
$\bar{\Delta} - 2.58\sigma > \Delta > \bar{\Delta} + 2.58\sigma$	________	________	$\leq P \leq$	________

REMARKS ____
____

Calculations by __________
Date ____________________

EXHIBIT 4A

QUALITY CONTROL DATA - ANALYSES OF SYNTHETIC SAMPLES
FOR PERIOD ____________________

				Bias				Precision, % (95% Confidence Interval)		Significance of Differences		
				Previous Quarter		Current Quarter				From Zero	From Previous Quarter	
Analysis and Method	Sample Identification	Unit of Measure	Standard Value	Arithmetic Amount	Percent of Mean	Arithmetic Amount	Percent of Mean	Previous Quarter	Current Quarter	Bias	Bias	Precision

EXHIBIT 4B

CONTROL LIMITS FOR ANALYSES OF SYNTHETIC SAMPLES
FOR PERIOD ____________________

(Based on Quality Control Data for Period ____________________)

Analysis and Method	Sample Identification	Unit of Measure	Control Chart Limits (95% Confidence Intervals)				
			Reported Value (Add to Standard Value)			Range in Replicates	
			Average	Minimum	Maximum	Average	Maximum

EXHIBIT 5A

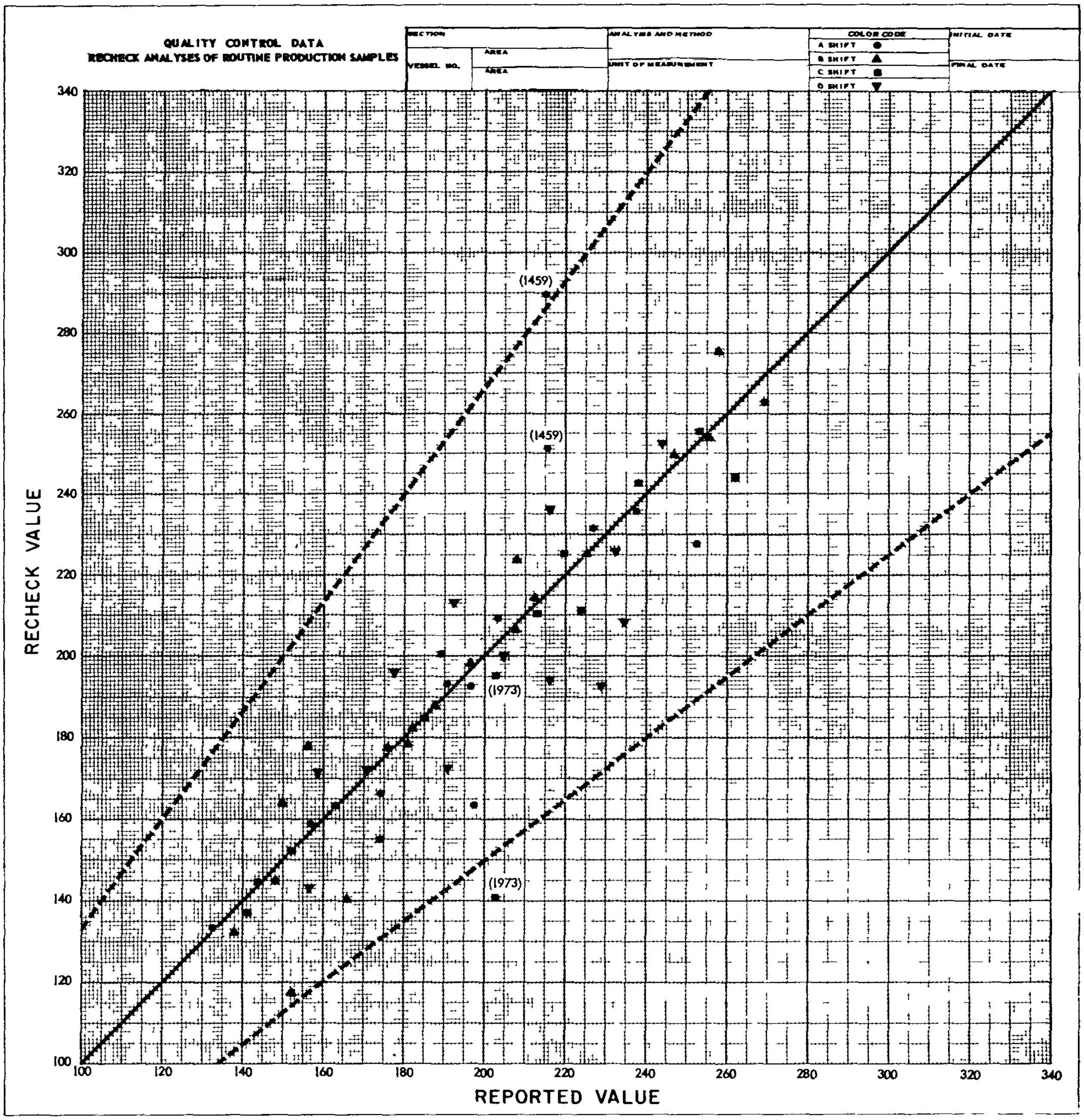

QUALITY CONTROL DATA
RECHECK ANALYSES OF ROUTINE PRODUCTION SAMPLES
SECTION
VESSEL NO.
AREA
AREA
ANALYSIS AND METHOD
UNIT OF MEASUREMENT
COLOR CODE
A SHIFT ●
B SHIFT ▲
C SHIFT ■
D SHIFT ▼
INITIAL DATE
FINAL DATE
RECHECK VALUE
340
320
300
280
260
240
220
200
180
160
140
120
100
REPORTED VALUE
100
120
140
160
180
200
220
240
260
280
300
320
340
(1459)
(1459)
(1973)
(1973)

EXHIBIT 5B

QUALITY CONTROL DATA

INITIAL DATE ____________

RECHECK ANALYSES OF ROUTINE PRODUCTION SAMPLES

SECTION

ANALYSIS AND METHOD

UNIT OF MEASUREMENT

VESSEL NO. — AREA

— AREA

IBM USE ONLY		
FIELD	IBM COL. NO.	CODE
A	1 – 2	1
B	3 – 4	
C	5 – 6	
D	7 – 9	
E	10	1

SHIFT	NO.
A	1
B	2
C	3
D	4
DAYS	5

FIELD	F			G	H	I	J	K	L
COL. NO.	11 – 16			17 – 21	22 – 26	27	28 – 32	33	34
LAB USE ONLY	DATE			SAMPLE NUMBER	REPORTED ANALYSIS		RECHECK ANALYSIS		IBM USE ONLY
I.D. NO.	MO.	DAY	YEAR		ANALYTICAL RESULT	SHIFT NO.	ANALYTICAL RESULT	SHIFT NO.	

EXHIBIT 6

QUALITY CONTROL DATA - DUPLICATE ANALYSES OF ROUTINE PRODUCTION SAMPLES FOR PERIOD ____________________

				Precision, % (95% Confidence Interval)		Significant Change in Precision Since Previous Quarter	
Analysis and Method	Tank No.	Unit of Measure	Median Value	Previous Quarter	Current Quarter	Better	Worse

ESTIMATING PRODUCT YIELDS AT HANFORD

GERALD R. PARKOS

Hanford Atomic Products Operation, General Electric Company,
Richland, Wash.

INTRODUCTION

Some of the factors which determine the methods used to compute product yields are: (1) The "regularity", or routineness and simplicity of the operation. (2) The availability and adequacy of machine computational facilities. (3) The state of the technology. Since these factors are always changing, the methods used to evaluate product yields must change correspondingly to keep up with the times. Several modifications have been, or are being, made to the techniques used at Hanford due to changes in the mode of operation, technological advances and improved computational facilities. This paper is devoted primarily to a general discussion of recent improvements in the methods used to compute product yields at Hanford.

ABSTRACT

The general concepts and methods used to develop yield equations are discussed briefly. Recent advances in nuclear technology, method of operation and computational facilities which have led to revisions in the methods used to evaluate product yields at Hanford are discussed. Among these are:

1. Adoption of the Westcott formulation for estimating nuclear cross sections.

2. Development of a versatile program for calculating product yields on the newly installed 709 computer.

3. Development of methods which will permit using the results of yield calculations to estimate exposure dependent reactivity parameters.

DISCUSSION

There are some very basic concepts and techniques which are always used in the development of yield equations. It is appropriate to review these concepts very briefly before discussing the specific techniques and equations used at Hanford.

Yield equations are always developed by coupling expressions (usually series expansions) for the isotopic concentrations as functions of exposure with the differential equations governing the rate of change of isotopic concentrations with exposure. For example, the Pu-239 concentration may be approximated by:

$$\text{Pu-239 concentration} = A_1 + A_2N + A_3N^2 + A_4N^3 + A_5N^4 + - - -$$

N = Fission concentration, the A's are appropriate constants.

The differential equation for the rate of change of Pu-239 concentration with fission concentration may be written in word form:

$$\frac{\text{Rate of change in Pu-239 con.}}{\text{Rate of change in fission con.}} =$$

$$\left[\frac{\text{Rate of change in 235 con.}}{\text{Rate of change in fission con.}}\right] \left[\frac{\text{Pu-239 producing neutron captures}}{\text{Neutron captures in 235}}\right.$$

$$\left. - \frac{\text{neutron captures in 239}}{\text{neutron captures in 235}}\right] = \left[\frac{\text{Rate of change in 235 concentration}}{\text{Rate of change in fission concentration}}\right]$$

$$\left[\frac{\text{non-resonance captures in 238}}{\text{captures in 235}} + \frac{\text{resonance captures in 238 from fissions in 235}}{\text{captures in 235}}\right.$$

$$+ \frac{\text{resonance captures in 238 from fissions in higher fissionable isotopes}}{\text{captures in 235}}$$

$$\left. - \frac{\text{captures in 239}}{\text{captures in 235}}\right]$$

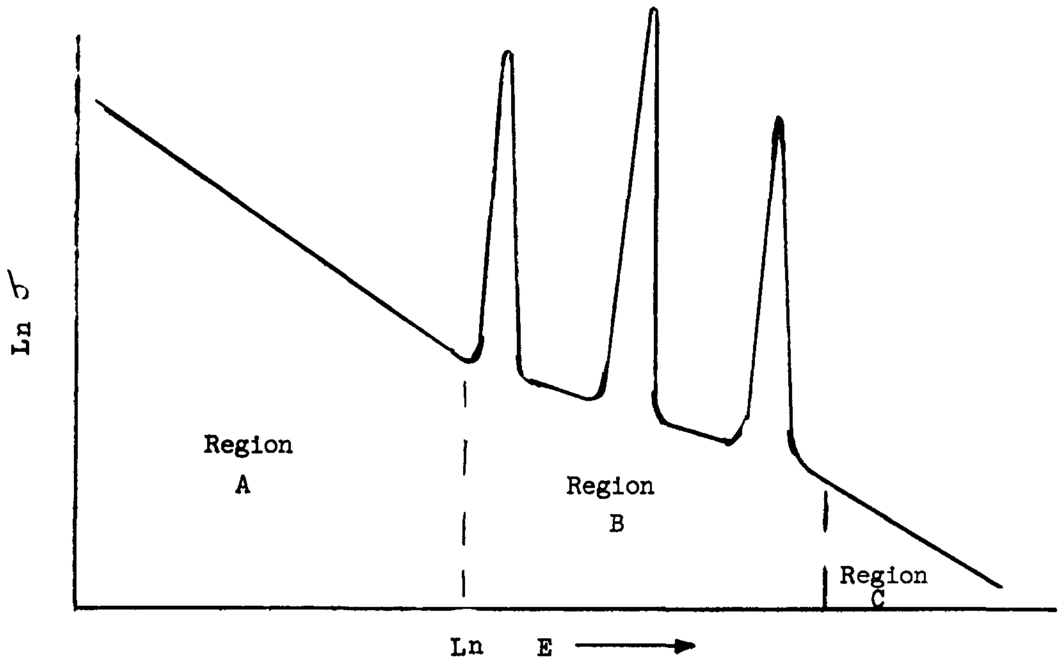
Similar differential equations and series expansions may be written for all of the isotopes of interest. The general method of solution for this type of problem is:

1. Write out the differential equations for all of the isotopes of interest.
2. Write out the expressions for the isotopic concentrations of interest.
3. Find the constants (A's in the example) in the expressions for isotopic concentration by solving between 1. and 2. above.
4. Use the resulting expressions for isotopic concentrations to estimate yields by substituting the appropriate nuclear and reactor constants and the known fission density.

The following items must be available before actual results can be obtained:

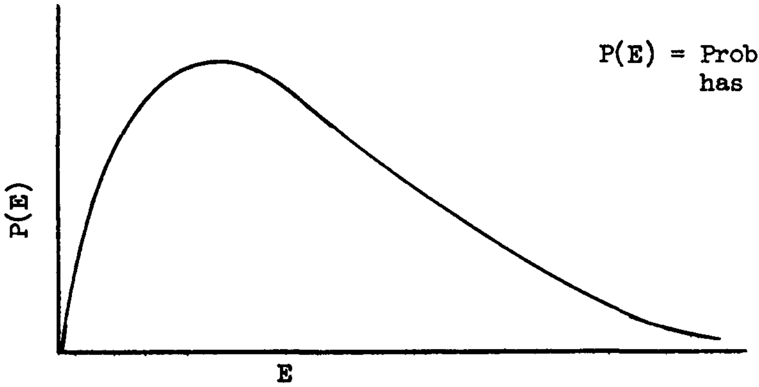
1. Nuclear cross sections.
2. The relationship between fission density and power density (the heat of fission).
3. The characteristics of the reactor (such as the resonance escape probability, leakage probability, neutron energy spectrum, etc.).

Approximations for neutron cross sections are always needed. At this point it is appropriate to discuss the approximations which have been made in the past and a fairly recent improvement (at least from the standpoint of method standardization). It is well known that the absorption cross section of a nucleus is a function of neutron energy. A general plot of nuclear cross section versus neutron energy might be drawn as follows:



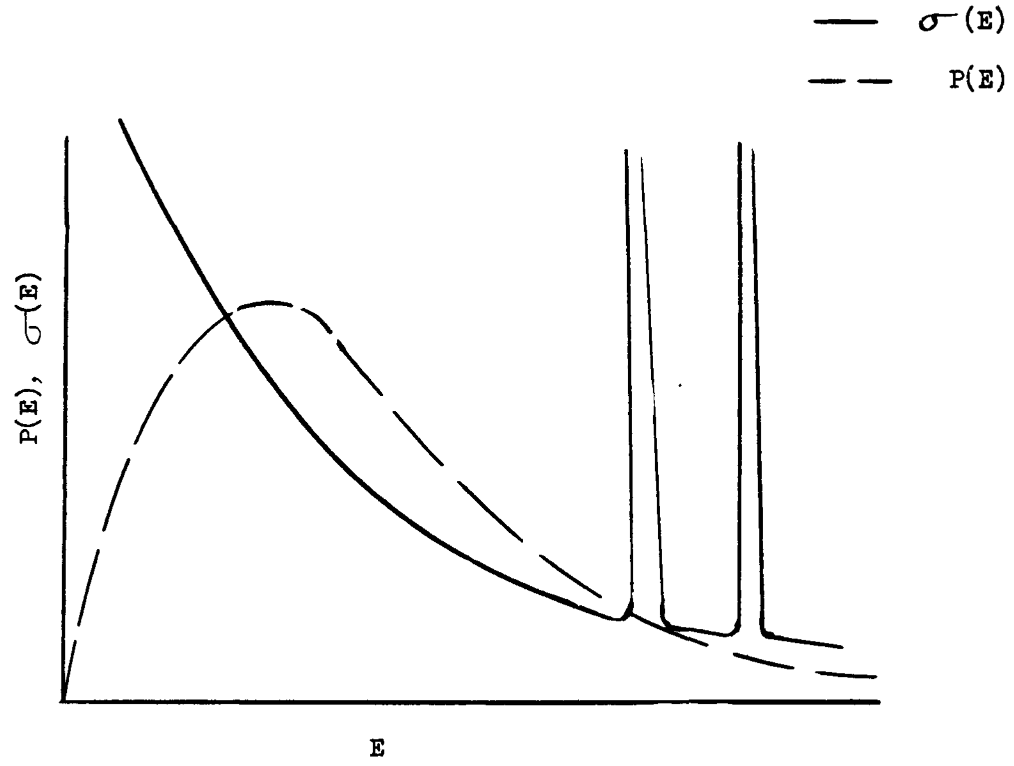
Region A is the "thermal" energy region. An ideal nucleus has a "1/v" cross section in this range. (That is the plot of ln σ vs. ln E is a straight line.) Region B is the "resonance" or "epithermal" energy region. Note the characteristic "resonance" peaks in the cross section curve. Region C is the "fast" neutron region. Note that the cross section is very small in this range.

The neutrons within a reactor have a range of energies rather than a fixed energy value. The neutron energy distribution in a thermal reactor is characterized by a Maxwellian distribution terminated by a 1/E tail. This may be represented pictorially as follows:



P(E) = Probability that a neutron has energy E.

Defining the representative cross section of a nucleus in a reactor is difficult. The problem may be seen pictorially by superimposing the nuclear cross section and neutron energy spectrum curves.



From this picture it may be seen that defining a single "effective" cross section for a nucleus is difficult even if the cross section and energy spectrum curves are completely known. Unfortunately, in many cases neither is known adequately.

The method used for determining the "effective" cross sections at Hanford, and most other installations in the past was:

1. Assume a Maxwellian distribution of neutron energies.
2. Correct the best available 2200 m/sec. cross section data to the temperature of interest assuming "1/v" behavior.
3. Correct this cross section for non-"1/v" behavior by the published correction factors.

This method is adequate for nuclei which do not have large characteristic resonances in the epithermal region. The resonances in U-238 were handled by using measured values of the "resonance escape probability". Most of the other resonances were ignored.

A method for representing cross sections proposed by Westcott[2] which has been widely accepted by reactor physicists has been adopted at Hanford. The Westcott formulation assumes that the neutron energy distribution is a Maxwellian terminated by a 1/E tail. The cross section of a nucleus is divided into two corresponding parts: (1) A thermal cross section characteristic of the nucleus of interest at the temperature of interest. (2) An epithermal cross section characteristic of the nucleus in the neutron energy distribution of interest. Thus, using this notation:

$\tilde{\sigma} = \sigma_0 (g + rS)$

$\tilde{\sigma}$ = Effective cross section

σ_0 = Neutron cross section at the appropriate temperature

g = Correction factor describing the non-"1/v"-ness of the cross section in the thermal energy region.

S = Correction factor describing the non-"1/v"-ness of the cross section in the epithermal region.

r = Epithermal ratio - A correction for the relative number of neutrons in the epithermal range.

The cross section obtained in this way includes a proper weighting for the epithermal resonances. This is particularly significant in product accounting because many of the Pu isotopes have large epithermal resonances.

At present the U-238 resonance captures are accommodated at Hanford by using measured values of the resonance escape probability. The Westcott formulation is used for all other nuclei. It is possible that the Westcott formulation may be used for U-238 in the near future.

Note that in order to use the Westcott formulation it is necessary to know the neutron energy spectrum. In particular, the thermal neutron "temperature" and the "epithermal" or "cadmium" ratio must be known. A method for calculating the "Epithermal Ratio" has been developed at Hanford.[3] Determining the proper thermal neutron temperature is, however, still a problem. The flux distribution, and hence the principal moderator (graphite) temperature, varies from point to point in the Hanford production reactors. Moderator temperature data cannot be used per se because:

1. The moderator and fuel elements are separated by a relatively low temperature water annulus. Since water is an excellent moderator, the water annulus reduces the effective thermal neutron temperature. The neutron "cooling" effect of the water annulus is not precisely known.

2. There is a good deal of theoretical and experimental evidence which indicates that the "effective" thermal neutron temperature is somewhat higher than the moderator temperature in a heterogeneous system. The reason for this is quite simple. Imagine the thermal neutron flux to be composed of "one pass" and "many pass" neutrons. ("One pass" refers to neutrons which make only one pass into the moderator, then are absorbed in the fuel. "Multiple pass" neutrons make more than one pass into the moderator before being absorbed in the fuel.) The "many pass" neutrons have reached thermal neutron energies. The "one pass" neutrons have a higher effective temperature because they have not reached thermal equilibrium with the moderator.[4] Therefore, the effective thermal neutron temperature is slightly higher than the moderator temperature.

Because of these difficulties the effective neutron temperature is determined indirectly at Hanford. Various neutron temperatures are used in the product equations, and the temperature which most nearly fits observed yield data is selected as the "effective" neutron temperature. It is possible that, by improving the technology, monitoring facilities and accounting techniques, some improvement can be made in this area.

The yield equations formerly used at Hanford were compressed (that is most of the higher order terms were eliminated) to permit easier use when manual calculations were necessary. This compressing did not result in a significant loss of accuracy but it did limit the useful range of the results. A new set of parametric differential equations has been developed, and the solutions to these equations have been programmed for the 709 computer. The program which has been, or is being, written will be versatile. It will permit computing product contents for a wide range of fuel element exposures and types.

The accurate evaluation of the isotopic content of irradiated fuel elements is of vital interest in reactor physics work because the neutron multiplying ability of a reactor is intimately related to the isotopic content of the fuel elements. Therefore, accurate yield calculations are of interest to reactor physics personnel because they permit estimating the variation of pile reactivity with fuel exposure. Estimates of the variation of the Hanford pile reactivities with fuel exposure have been based primarily upon experimental tests in the past. These tests consisted of reactivity measurements at the production reactors, and in test assemblies. It is quite probable that much more emphasis will be placed upon theoretical calculations (direct use of yield calculations) in the future because:

1. The mode of operation of the production reactors is becoming more complicated and streamlined all the time. Therefore, time for experiments is hard to get, and such time is very costly when it is available.

2. It is very difficult and costly to perform adequate test pile experiments. Mocking actual reactor conditions in a test assembly often borders on the impossible.

3. Theoretical calculations can provide reasonably accurate results.

This is a healthy situation from the product accounting viewpoint because it means that more people will become more interested in obtaining reliable yield equations.

Reactor physics personnel are designing tests which will provide accurate determinations of product yield and composition as functions of fuel exposure. It is premature to speculate on what kind of tests will be conducted. However, current thinking indicates that they probably will be similar to the tests described in Reference 1. These tests consisted of irradiating fuel elements to various exposures in an accurately known flux. The elements were then analyzed for isotopic content using a mass spectrometer. The reactivity of the elements was measured in a test assembly. It is felt that this kind of test is superior to tests involving accurate separations plant determinations of the product content of large batches of elements because:

1. Separations plant uncertainties are eliminated.

2. Small scale tests do not affect reactor efficiencies significantly.

3. Small scale tests can provide vital physics information which often leads to major advances in techniques and knowledge. Batch type tests usually provide data which permits "fudging" the constants in the yield equations to fit observed results. Such data usually cannot be extrapolated beyond the range of the experiment.

Up to this point nothing has been said about the reactor data actually used to compute the product content of irradiated fuel elements at Hanford. The power of individual fuel channels is monitored routinely by Operations personnel. Representative maps of the radial flux distribution, automatically printed or taped, are machine analyzed by Production Scheduling Operation. Production Scheduling Operation keeps a running account of the product status of each fuel channel, and schedules them for discharge at the appropriate time. The exposure data are fed into the product yield equations discussed previously, and the product content is computed by machine on an individual channel basis. This is an improvement over previous methods where the product content of a group of fuel element channels was estimated on a "batch" basis.

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RECOVERY OF SOURCE AND SPECIAL NUCLEAR MATERIALS

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ABSTRACT

The procedures for handling scrap SS materials which have been transferred to the UCNC Y-12 Plant for recovery are discussed. The application of proper segregation and packaging of such scrap to economical processing is illustrated by typical process flows. A few of the problems associated with the evaluation of scrap SS material are mentioned.

INTRODUCTION

A sizeable part of the Y-12 Plant operations, from its early days as an electromagnetic separation plant to the present time, has been concerned with the recovery of uranium from various internally generated scrap and waste materials. Today in addition to the internal program, a substantial amount of recovery is done for other AEC contractors. This experience with our own materials has made us very familiar with the problems of the generating contractor faced for the first time with waste which combines the usual chemical hazards of toxicity, corrosion, and explosion with potential radiation dangers.

The fact that there was available in Y-12 a large and efficient recovery plant for unirradiated uranium scrap of most assay ranges led to the utilization of these facilities in processing salvage for other facilities, first from the other UCNC plants in Oak Ridge and later from practically all other AEC contractor generators of this type of material. It was soon recognized that the nonuniformity of scrap shapes, the wide range of concentrations, the variations in contaminants, and the unlikely combinations of materials received from other contractors would serve to compound the usual chemical process difficulties encountered in our own recovery operations. As a result, special procedures were developed, which supplement the routine SS control methods, and provide special handling for all scrap shipments from the time a shipment is received until the SS material amounts recovered are reconciled with shipper's values. At the same time, a need was indicated for standardization of shipping and handling methods, as well as a means of classifying the various forms of scrap in accordance with the processing operations required for recovery.

The development of standard procedures for recoverable scrap has been a long and sometimes tedious process. No simple definition or guide could begin to consider all of the difficulties encountered by a recovery plant in evaluating, measuring, and accounting for the contained SS materials. In addition, there are unique problems associated with the accumulation, packaging, transporting and processing of these materials.

The development of answers to many of these problems and the final issuance of Chapter 7430 has been of great satisfaction to Y-12 personnel. We look upon it as a major milestone in the improvement of scrap recovery. Even though further amplification and revision will undoubtedly be necessary, the availability of this guide should be of assistance to all handlers of scrap containing SS materials for recovery.

RECEIVING SS SCRAP

All scrap for recovery at Y-12 is received by SS Control personnel in a designated security area. Before any shipment is unloaded, the shipping papers are reviewed by a responsible person for obvious irregularities in classification, weights, or packaging. All containers are then gross weighed and checked against the gross weights listed on the AEC 101 Form or attached tally sheet. A Form AEC 284 is then prepared for each shipment and a copy returned to the shipper acknowledging receipt of the gross shipment in the SS Control warehouse. A cursory evaluation is also made at this time to determine the need for special storage precautions and to disclose material of borderline recovery value.

All SS materials handled at Y-12 must meet certain minimum conditions of physical and nuclear safety as well as standard chemical toxicity regulations. Specialized groups directly concerned have established adequate procedural safeguards. For example, Y-12 nuclear safety standards require the parameters for salvage storage shown in Table I. All storage of enhanced uranium must conform to these regulations. The use of containers mentioned in the table and recommended in Chapter 7430 is not a rigid requirement but deviations present serious storage problems and should be made only after consultation with the recovery contractor. A scrap material shipping guide approved by the Y-12 nuclear safety group is shown as Table II.

The dead storage time for any shipment is dependent upon the backlog of SS material for recovery. Economic operation places a priority on the recovery of scrap which contains the greatest equity of SS material which can be recovered at the least cost. Consequently, combustibles and other scrap with low SS material equity may be in storage for an extended period. A status report of all shipments awaiting recovery is routinely prepared as an aid to efficient scheduling of production. The campaign processing of a number of shipments of like materials from the same shipper is done whenever practical.

PRE-MEASUREMENT PROCESSING

The pre-measurement treatment of scrap at Y-12 is accomplished by special operations groups. The identity of each shipment is maintained as it is transferred from the SS Control warehouse to the appropriate operations area for checkweighing of each container and further classification for processing.

TABLE I

PARAMETERS FOR Y-12 SALVAGE STORAGE[a]

Type Container	Mass Limit (U-235)[b]	Comments	Area Limits[c]	Category[d]
Standard Birdcage[e]	< 18 kg		Maximum of 40 cages in contact/array	A, B, D
Slotted Angle Type Birdcage[f]	2 kg	Inner container volume must be < 4 liters	Maximum of 40 cages in contact/array	D, C, E
Special Drum[g] Solution Container	~ 5 kg[h]	Inner container 5-1/8" I.D.	4/pallet, 3 x 8 pallet array	E
Special Drum[g] Scrap Container	2 kg	Inner container 6" I.D.	4/pallet, 3 x 8 pallet array	C
55 Gallon Drum	≤ 350 gms	--	4/pallet, 3 x 8 pallet array	E, F, G, H
	≤ 100 gms	Specific Application	Any convenient array	
30 Gallon Drum	≤ 350 gms	--	4/pallet, 3 x 8 pallet array	E, F, G, H
	≤ 100 gms	Specific Application	Any convenient array	
Other Containers[h] (> than one gallon)	200-350 gms	--	One diameter separation. 4/pallet, 3 x 8 pallet array	C, D, E, H
	≤ 200 gms	--	1 kg/pallet[i], 3 x 8 pallet array or 100 gms/sq ft shelf[j]	
Other Containers (one gallon or less)	< 350 gms	--	100 gms/sq ft shelf[j]	C,D,E, H
MAC Special 20" Container	< 500 gms	One container per 20" box	2 kg/pallet, 3 x 8 pallet array	C

(a) Due to the complexity of the critical variables, a simple table cannot outline all safe limits. Although nonconformities to this table may be safe, each must have a special evaluation.

(b) When Be or D_2O are contaminants, special evaluation is required.

(c) There should be a 4 ft minimum separation between all arrays.

(d) AEC Chapter 7430-033B-2.

(e) Standard birdcage provides a minimum center-to-center separation of 20".

(f) This may be a single or multiple unit cage. It must provide a minimum of 20" center-to-center separation between units.

(g) Modified 55 gallon drums.

(h) About 5 kg is maximum for optimum solution at H:X = 45. Any outside container that provides the same separation as a 55 gallon drum has the same limits.

(i) Whenever possible, maximum separation should be utilized.

(j) Shelves are on a minimum 18" vertical separation.

TABLE II

SCRAP MATERIAL SHIPPING INFORMATION

Description (Category*)	Physical Condition	U-235 Mass†	Type Container
Uranium Metal or Alloys (a,b,c)	Solid Masses	< 18.5 kg	Standard 20" Birdcage‡
Uranium Compounds (d)	Oxide or Salts—Dry	< 18.5 kg	Standard 20" Birdcage‡
Uranium Metal or Alloys (c)	Foil, Fines, or Turnings—Under Oil	< 2 kg	Standard 20" Birdcage or Special Drum Scrap Container§
Fuel Elements (a,b,c)	Metal Pieces—Dry	< 2 kg	Standard 20" Birdcage or Special Drum Scrap Container§
Heterogeneous Mixtures (c,e)	Slag, Fines—Wet or Dry	< 2 kg	Standard 20" Birdcage or Special Drum Scrap Container§
Processing Solutions (e)	Concentrated Solutions	< 2 g/l	Standard 20" Birdcage or Special Drum Solution Container§
Salvage Solutions (e)	Dilute Solutions	< 2 g/l	55-gallon Drum
Combustibles (f)	Miscellaneous—Dry	<250 g	55-gallon Drum
Noncombustibles (g)	Miscellaneous—Dry	<250 g	55-gallon Drum
Residues (h)	Miscellaneous—Wet or Dry	<200 g	55-gallon Drum

* Letters in parentheses refer to AEC Chapter 7430-033B-2.

† When Be or D_2O are contaminants, other criteria may apply.

‡ Maximum of 50 cages in contact or array.

§ Maximum of 60 special drum containers in two-dimensional array.

Source: Guide to Shipment of U-235 Enriched Uranium Materials, TID 7019.

In Figure 1 we have illustrated the various steps followed by every shipment of scrap from its receipt until shippers' and receivers' measurements are compared and reconciled. It will be noted that sampling and analysis is done at the first opportunity. Unfortunately, the majority of the material is of such nature that extensive physical or chemical treatment is required before reliable measurements can be made.

Proper segregation of scrap is very important. If scrap is to be processed economically, it must be classified according to the specific operations required to bring it to a measurable state. Therefore, group classification and batching for nuclear safety are among the first process steps. Each type of material requires different preliminary treatment. If proper segregation is made by the shipper, and separate shipping forms prepared for each class, both processing and proper accountability will be expedited.

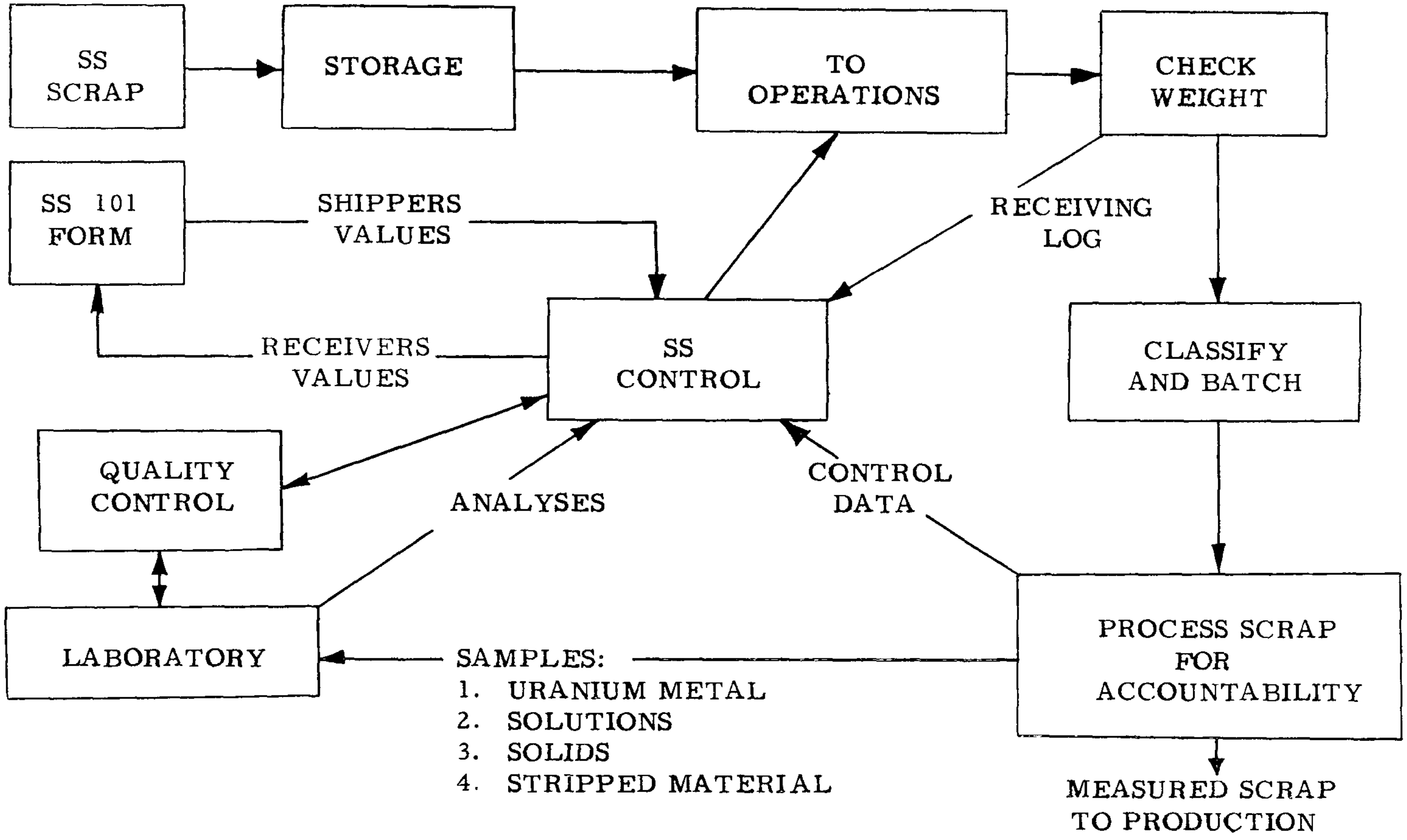

Fig. 1—SS scrap receipts at Y-12.

In all cases segregation is based upon the specific process requirements of the material. Materials are grouped for processing as shown in Table III.

TABLE III

MAJOR GROUPS BY ACCOUNTABILITY PROCESS

Group 1.	Recastable U metal, machine turnings and non-combustibles that require degreasing or pickling.
Group 2.	Various uranium compounds, alloys and other SS materials that may be directly dissolved.
Group 3.	Aqueous and organic solutions that require only filtration.
Group 4.	Combustibles of all classes that must be burned.
Group 5.	Ash and residues that must be dried and milled.
Group 6.	Miscellaneous materials.

The process flows necessary to bring each of the above groups to a measurable state are illustrated in Figure 2. It is immediately apparent that the materials in Group I represent much less of a processing problem than those in Group II.

What may not be so readily apparent is the complexity of the operations that may take place in each of the illustrated steps. The dissolving of various types of material can be done in several ways, each requiring a different procedure. Examples of common types and four of the usual dissolution methods are shown as Table IV.

TABLE IV

DISSOLUTION CLASSIFICATION

Examples	Scrap Classification*	Method of Dissolution
UO_2 - U_3O_8	EE	1. HNO_3
MgO Ceramics	HE	
SS Alloy	CE	2. Electrolytic
Zr Alloy	CE	3. HF and HNO_3
Zr Turnings	DE	
SS Turnings	DE	
ThO Ceramics	HE	
Al Alloy	CE	4. Caustic
Al Turnings	DE	

* AEC Chapter 7430.

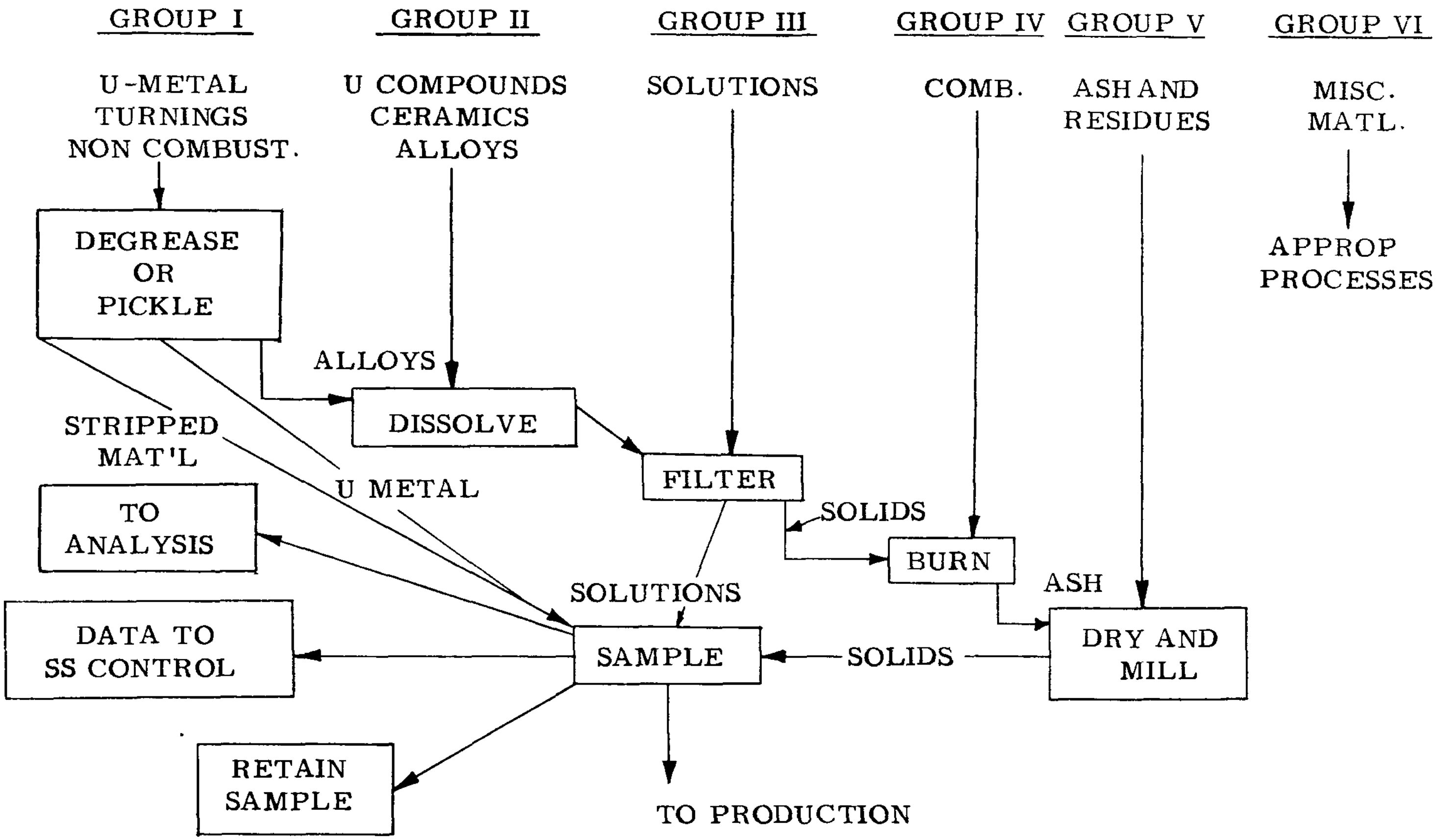

Fig. 2—SS scrap recovery to accountability.

The particular method used is dependent on the class and condition of the scrap, with specific handling and batching problems presented by every type of material. Normally each container is checkweighed as received and assigned a control batch card to provide identification for processing and nuclear safety. This control is continued until the material is sampled at which time an SS Control batch number is assigned. However, exceptions may be made as in the case of zirconium alloy turnings where accurate checkweighing is not feasible due to the nature of the material. In this case batching is done on a volume basis.

It may also be necessary at times to compromise the identity of quota assignments or individual containers in the interests of processing and sampling economy. For example, the dissolving of uranium aluminum alloy elements and scrap is very complex and is not adapted to batch processing. In this or any other continuous system, it is very desirable to combine shipments of the same type material for processing. The shipper is given credit for all product and waste created during a campaign. In the lower U-235 concentration ranges, the campaign philosophy is used very frequently.

It is desirable both from the shippers' and receivers' point of view to keep the amount of pre-measurement processing to a minimum and to sample every shipment at the first opportunity. The exact method of measurement will depend upon the physical condition of the material, but, in every case, either the sampled material or a generous retain is held until analytical results have been evaluated. The Y-12 Quality Control group maintains close check on analytical methods and, in addition, spikes duplicate samples from at least 10% of the batches in each highly enriched shipment. Any shipper-receiver discrepancy is resolved in co-operation with this group and re-samples submitted if necessary. The shipping station is then credited with the proper SS value and further processing of the scrap takes place in the regular production recovery stream.

URANIUM RECOVERY

The four common types of measured material arising from the pre-measurement treatments just discussed are introduced to appropriate points in the main recovery cycle as illustrated in Figure 3. It becomes immediately apparent that metal, for example, is much easier and more economical to recover than milled solids. Normally, pure metal either in the form of turnings or massive scrap will be in the main process stream in a matter of days.

Solutions for recovery will usually require adjustment of acidity and specific gravity prior to the extraction and purification steps. It should be pointed out that most of the highly enriched uranium is extracted from nitric acid solutions in a stainless steel extraction system utilizing di-butyl carbitol as the organic and that the presence of any halide is detrimental. It is possible to complex the fluoride ions found in many pickle solutions but the removal of chloride ions, requires extensive and costly pre-extraction treatment.

The purified uranyl nitrate obtained from the extraction operation is converted to UF_4, with HF, either in a continuous fluid bed or batch conversion reactor. An ion exchange system is utilized for the extraction of slightly enriched materials in a sulphate medium.

The additional leaching and residue treatment required by solid material add materially to the recovery problems and costs. The leachates from this process are treated as solutions while the leached residues are processed to a point where further treatment is not economical. They are then measured for discard and removed from inventory in conformance with AEC Procedures.

All raffinates and condensates arising from the main recovery process are likewise measured for discard on the same basis. Recovery efficiencies of greater than 99.9% are usually experienced in highly enriched material with efficiencies in the lower ranges commensurate with the contained uranium values.

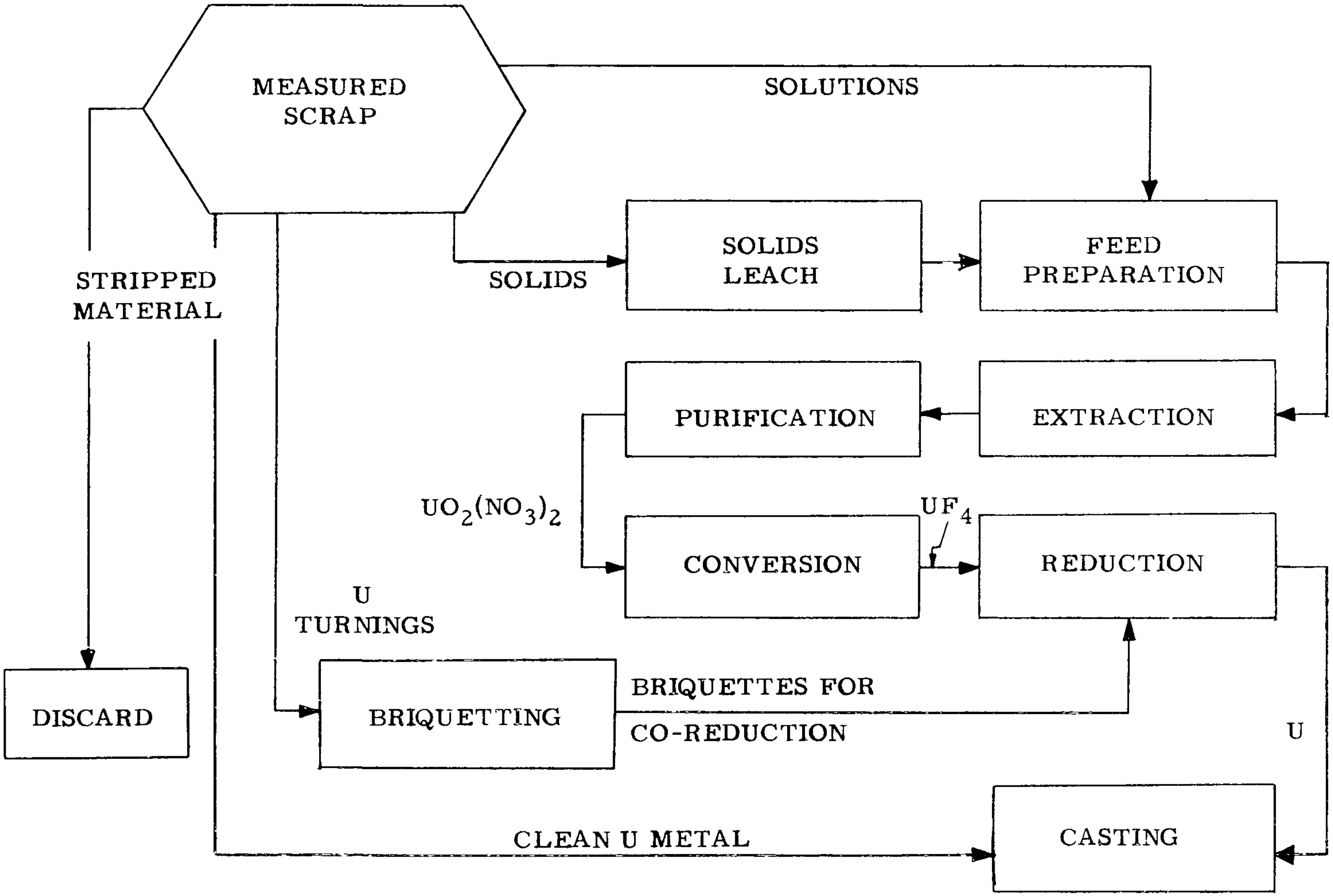

Fig. 3—SS recovery to production channels.

The standard recovery processes at Y-12 produce final compounds of uranium with average impurities of < .2%. The impurities in intermediate compounds and cast metal, may be slightly greater.

EVALUATION OF SCRAP

One of the basic questions facing every generator of scrap is how to best handle his waste material so that economies in operation are not offset by increased recovery costs. An evaluation of each type of scrap material produced is therefore necessary.

There is really no question that classes AE through FE described in Chapter 7430 are economically feasible to process when the total U content is greater than 0.1%. However, the feasibility of recovery of classes GE, HE, and JE is not so straightforward.

To help illustrate this point let us take for example a mythical shipment of combustibles from Station XYZ. This station had only recently become engaged in the fabrication of fuel elements. Recognizing the extreme value of their waste streams, supervisors had instructed their people to collect all floor sweepings, gloves, wipes, and other contaminated combustibles for possible recovery. It had been a practice at this plant to dispose of waste directly from the collection cans into a large garbage disposal unit by the janitorial forces and so new collection procedures were developed.
The collection of contaminated waste was delegated to a zealous young engineer who had been assigned two of the janitors. His first concern was shipping containers. Now this plant had regularly available from another department empty drums of 10 gallon, 30 gallon, and 55 gallon capacity and so it seemed only good business to utilize these containers for the collection and shipment of the new type waste.

As the first production day dawned tension had mounted and everyone in management was concerned with the U concentration of the fuel, the machining characteristics of the alloy or the accountability for the finished elements. The janitors who had been properly impressed had carefully numbered the variegated empty drums and, as the waste containers were filled, transferred their contents for shipment to the recovery plant. Now in this plant, as in most plants, the handling of the massive scrap and machine turnings had been given enough consideration to prevent serious handling or measurement difficulty. However, "combustible waste" is a broad term and habit is a strong factor. So it was not strange to find in drums of combustibles a wide variety of material ranging from uncontaminated chewing gum wrappers through gloves, wipes, oily rags, and, on occasion, machine turnings and slugs. Also, the use of non-standard containers allowed substantial variation in bulk density making comparison from shipment to shipment next to impossible.

This example is not intended as criticism, for it is really gratifying to report that most of the scrap received at Y-12 for processing has reflected the sincere efforts of the generating contractor to segregate material for recovery properly. It does illustrate that it is within his own operations that the scrap generator can, by proper evaluation of his waste streams, initiate greatest recovery savings. Proper organization may allow the measurements of the waste streams of greatest equity to be sufficiently accurate so as to preclude the necessity of processing combustibles and other difficult-to-handle residues. A better appreciation of the recovery processor's problem should also help to place the proper emphasis on the handling of all wastes.

UNIT COSTS

The evaluation of the feasibility of processing particular shipments is not a new problem at Y-12. For a number of years, we have been faced with the determination of whether the uranium contained in our own waste materials was economical to recover.

Recently a standard cost catalog (Y-1247) has been issued which makes available the approximate costs per unit of bulk for a large number of scrap materials. The development of unit costs were affected by operational problems peculiar to recovery processes. First, the cost of processing to a sample point is affected not only by the number and size of containers in the shipment but also by the exact condition and concentration of the scrap therein. Second, the costs of recovery from a measurement point are more directly related to weights and volumes of material processed than to uranium content because of the continuous nature of the operation. Third, any costs required to reconcile shipper-receiver differences has to be included.

The application of bulk unit costs for evaluation purposes is relatively simple. If the uranium content of a shipment is well known, the processing cost per gram of U can be determined by utilization of the Y-12 costs in the following formula:

$$\frac{\text{Cost of Processing to Sample Point + Cost of Sampling and Analysis + Cost of Recovery}}{\text{Estimated U Content (grams)}} = \frac{\text{Processing Cost}}{\text{Per Gram U}}$$

If the uranium content of a specific shipment is not well known, it is possible to determine the minimum uranium content per unit of volume that is economical to recover. For example:

$$\frac{\text{Cost of Processing X drums combustible waste}}{\text{Value of U per gram}} = \frac{\text{Minimum grams U for}}{\text{Feasible Recovery}}$$

Thus if the generating contractor is in a position to state the uranium content within close limits for a particular type of scrap a better evaluation can be made.

The unit costs now available are based on past experience. It is anticipated that improvements in waste handling techniques and recovery methods will allow reductions in these amounts. Thus uranium contained in scrap residues which are not currently feasible to process may be recoverable at some future date. Provision for the stockpiling of this type material has been made in Chapter 7430.

CONCLUSIONS

In conclusion, we feel that the problems facing every generator of scrap are very similar to those experienced over the years at Y-12 in the generation and collection of its own scrap. The early adaptation and application of Chapter 7430 to each contractor's specific problem should result in a safer and more economic operation. The standardization of types and classes of material and the evaluation of process waste streams at their source should reflect a substantial reduction in the problems encountered by the recovery contractor and result in lower and more stable recovery costs.

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APPLICATION OF A RATIO METHOD OF PLUTONIUM RECEIPT MEASUREMENT IN A CONTINUOUS SEPARATIONS PLANT

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INTRODUCTION

The criteria for a fully adequate separations plant control system requires the consideration of the same fundamentals that are common to any process subjected to material balance techniques. The Hanford separations plants receive irradiated normal solid slugs, canned in aluminum. Plutonium and uranium are decontaminated and separated. Input quantities, of course, are a dominating factor in the material balance. A good receipt measurement is therefore required as the starting point for satisfactory control. The term good is purely qualitative, but to put it in quantitative terms, a measurement as good as or better than output measurement. If such a measurement is not available, it is impossible to distinquish between pile and separations plant measurement uncertainties, or between measurement uncertainties and loss of material. Such a status prevents competent evaluation and is inadequate for material balance control.

For many years a receipt measurement has been made on dissolver solution using standard sampling, volume, and analytical techniques. All three phases have contributed uncertainty to an extent which made the measurement little if any more reliable than the pile production estimate.

As recently as a year ago it was felt the problem could be resolved only by conducting closely controlled transmutation tests which could firmly set calculation constants for each pile, type of material, and location in the pile; thereby firming up the pile calculations. Accurate dissolver solution measurement was not deemed possible, partly for technological reasons and partly for economic considerations. A task force was assigned the task of working out the details for special transmutation tests. The most accurate measurement found for these tests was one based on acceptance of uranium weight and determining a plutonium to uranium ratio, thereby arriving at a plutonium value essentially independent of sampling and volume considerations. It was found before tests were started that it was possible to incorporate the ratio type measurement on a routine basis to measure all separations plant receipts.

This paper discusses events leading up to adoption of a ratio method for routine receipt measurement, the details of its application in a Purex continuous process, method obstacles and their elimination, and the advantages to be realized with the ratio type measurement.

EVENTS LEADING UP TO ADOPTION OF A ROUTINE RATIO METHOD

The starting point for the HAPO chemical processing material balance is the pile estimate value. Error in these calculated values have caused B-PID's in chemical processing plants. Experience has shown there have been periods of overstatement and periods of understatement with overstatements predominating in recent years. The many changes in pile operating conditions, fuel element design, and adding new piles have made many pile production calculation changes necessary. Conversion constants have been changed on a theoretical basis whenever a pile physical change was made. Although chemical processing data at times confirmed changes made, there has been no direct use of processing data for determining calculation constants. The lack of agreement in chemical processing measurements at various points in the processing has contributed uncertainty as to the ability of the separations plant to measure receipts. For this reason, it has not been possible in the past to ascertain when pile calculations require refinement; and whether B-PID is the result of pile calculations or the result of separation operations. There are three points in the separations area at which comparisons can be made to pile statements; (1) dissolver solution measurement, (2) separations plant loadout, and (3) finished product loadout. In each case the comparison is based on a material balance up to that point which includes much inventory and as a result much added uncertainty.

1. The Dissolver Solution Measurement

This measurement is the least reliable because it requires a volume determination and sampling must be done remotely using extremely small samples. The measurement, as with all three points, is limited by the inventory uncertainty up to the point at which the measurement is made.

The reliability of this measurement as a receipt verification is no better than ±6 per cent on the monthly basis and ±2 per cent on the yearly basis.

2. Separations Plant Loadout

Individual batch measurement precision is estimated at ±0.73 per cent and is now believed to be unbiased. As a comparative point for verification of pile receipts, this measurement itself is quite accurate but has a greater inventory drawback than the dissolver solution point since the entire plant inventory is involved in striking up the necessary material balance for the comparison. Compounding the potential error up to this point are the added waste streams in the balance.

On the monthly basis the verification is probably good to ±5.0 per cent and to ±0.4 per cent on a yearly basis.

3. Finished Product Loadout

The accuracy of finished product measurement has always been good and contributes essentially no uncertainty to the control. However, using the material balance up to this point as a comparison of pile measurements has little significance because of the uncertainty in inventory up to this point. On a monthly basis the comparison is good only to about ±10.0 per cent and on the yearly basis to ±3.0 per cent.

In the Fall of 1957 it was concluded that accurate measurement of all receipts was not at present economical, but it did seem possible to conduct special accurate measurements on a small number of closely controlled irradiations to permit improving the accuracy of the pile calculations. A task force was formed to outline the requirements of such a program. A ratio method for plutonium measurement was recommended by the Process Chemistry Operation as the measurement giving the greatest accuracy. The method was based on acceptance of pile uranium as an accurately measured quantity, and the analytical determination of the Pu/U ratio in the dissolved solution. An accuracy of ±0.4 per cent per measurement was believed possible. Once all the details were worked out and test procedures written, it was realized that cost of the program would be excessive particularly in the pile areas where segregation required down time. The extreme cost and the fact that tightly controlled tests may not be representative of normal pile measurements caused interest in this program to dwindle. The fact remained that the only true measure of pile calculation accuracy is an accurate 100 per cent measurement at the separations plant.

With the realization that only 100 per cent measurement would provide the needed control, a way was sought to utilize the proposed ratio method on all receipts. At first look it would appear this could be done only on a batch basis, definitely identifying a specific charge with its ratio after dissolution. This is not possible since heels must be maintained in dissolvers to increase their capacities. It was found, however, that a ratio determined at the routine dissolver solution measurement point could be used, quite accurately, to determine plutonium received on a longer term basis, of a month or more.

THE RATIO METHOD OF PLUTONIUM RECEIPT MEASUREMENT

The ratio method is based on the assumption that the pile uranium statements are essentially free of error. At the present time HAPO pile uranium statements are believed accurate to within ±0.1 per cent on the long term.

Knowing the uranium quantity the plutonium can be measured by determining the ratio of plutonium/uranium. In obtaining the ratio, the only error contributed would be that of an analytical nature. Possible sampling error would be minimized since dilution or concentration will not change the ratio. No gross measurement such as volume or weight is involved, other than to assure proper weighting of each batch analytical result in the final ratio. Thus the over-all measurement error potential is reduced to a minimum. An accuracy of 0.5 per cent should be possible on a monthly basis and approximately 0.10 per cent on the yearly basis. The monthly error is essentially that contributed by analytical. On the yearly basis the measurement accuracy approaches that of the uranium statements since analytical bias is measured and corrected for. It is realized that complete elimination of bias is wishful thinking, but since all known potential analytical bias is controlled, long term analytical bias will be small.

METHOD OF APPLICATION IN THE CONTINUOUS PUREX PROCESS

A measurement cannot be made on an individual charge basis for two reasons. First, the dissolvers must retain a heel to speed dissolving and increase capacity. Second, individual charges do not always carry an accurate uranium weight due to mixtures of piece type and the weight averaging system used.

The ratio *can* be applied on a monthly basis for determining a weighted ratio downstream at the dissolver solution measurement point, and applying it to the total month's uranium charge.

Figure 1 shows roughly the mechanics of the application.

Immediately, it would appear obvious that some accuracy would be lost by determining a ratio at one point and applying it at another. However, the only accuracy lost due to this lag is on the short term because the inventory in between charge point and ratio point will be small compared to a month's throughput. On the yearly basis the lag inventory will be insignificant. Actually it is only when the ratio of Pu/U in the inventory is grossly different from the month's average that any significant error is involved in the month calculation. Monthly errors cannot be additive if a cumulative measurement calculation is used as shown in Fig. 2. A more detailed cumulative system shown in Fig. 3 will be discussed later. Its purpose is to prevent error due to non-weighted use of the isotope correction factor.

At the E-5 dissolver solution measurement point, measurement is on a batch basis so it is possible to obtain a 100 per cent weighted ratio on the month's throughput by individually measuring plutonium and uranium per batch. As shown in Fig. 2, corrections are made to gross measurements for analytical bias as measured in the analytical quality control program which includes americium and curium in the case of plutonium, and for any recycle or other material which is not virgin metal but measured through this point.

Before discussing the potential causes of error in the specific phases of this ratio measurement, if we briefly go through the monthly calculation worksheet to observe the mechanics involved, you will observe where potential error may exist.

Note Fig. 2. The gross dissolver solution measurements for uranium and plutonium (which includes all three measurement phases, volume, sampling, and analytical) are still used to give proper weighting to each sample in the ratio calculation. Possible volume and sampling

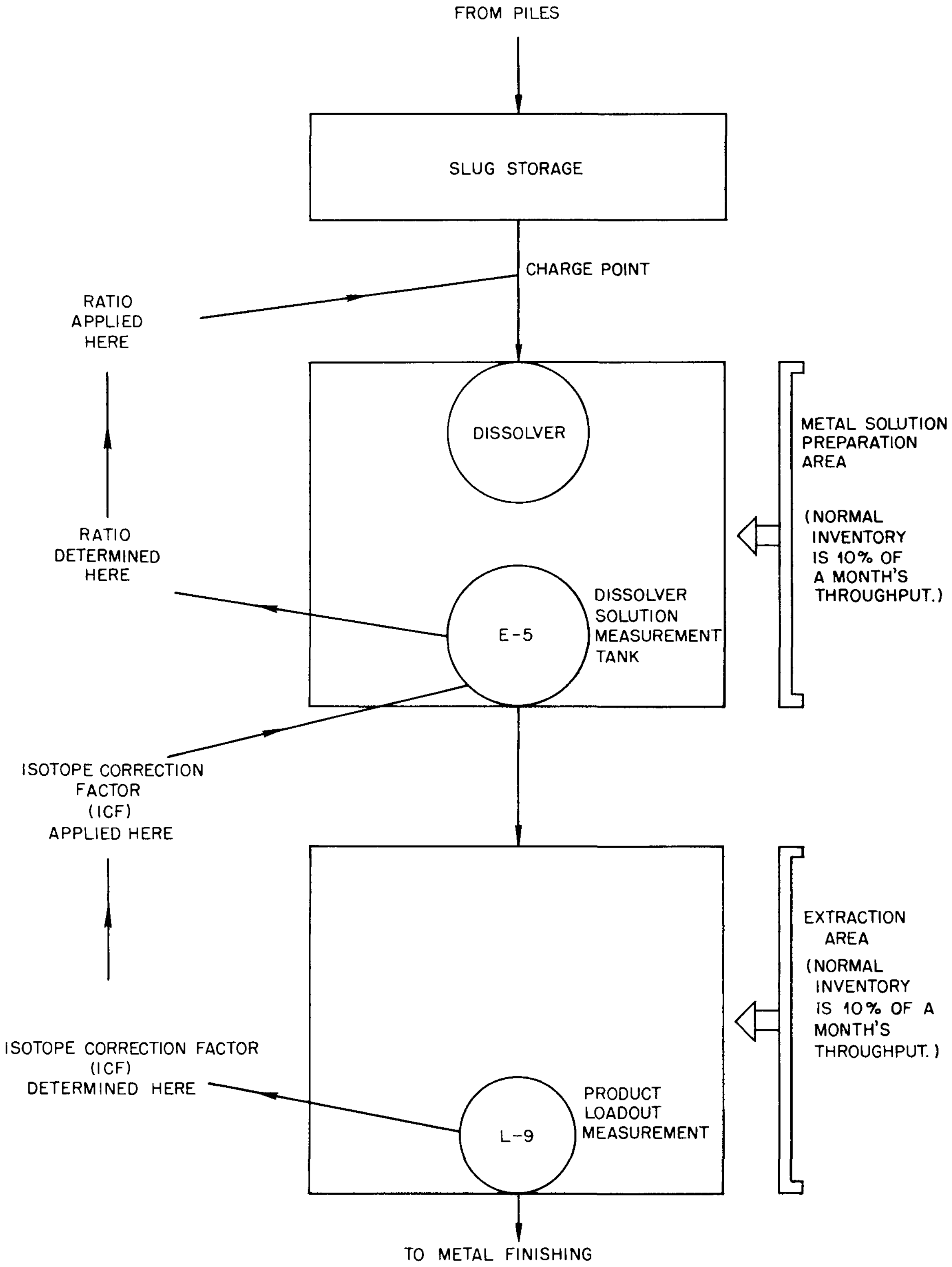
FROM PILES
SLUG STORAGE
CHARGE POINT
RATIO APPLIED HERE
DISSOLVER
RATIO DETERMINED HERE
E-5
DISSOLVER SOLUTION MEASUREMENT TANK
METAL SOLUTION PREPARATION AREA
(NORMAL INVENTORY IS 10% OF A MONTH'S THROUGHPUT.)
ISOTOPE CORRECTION FACTOR (ICF) APPLIED HERE
EXTRACTION AREA
(NORMAL INVENTORY IS 10% OF A MONTH'S THROUGHPUT.)
ISOTOPE CORRECTION FACTOR (ICF) DETERMINED HERE
L-9
PRODUCT LOADOUT MEASUREMENT
TO METAL FINISHING

Figure 1

RATIO METHOD
PLUTONIUM CHARGED TO DISSOLVERS
WORK SHEET

A. Current Month Virgin Plutonium Through the Dissolver Solution Tank
  (1) Gross plutonium removed
  (2) Plutonium total analytical bias as a % recovery
  (3) Recycle

$$\underset{(1)}{(\underline{\qquad})} \times \underset{(2)}{\left(\frac{100}{\underline{\qquad}}\right)} - \underset{(3)}{(\underline{\qquad})} = \underset{(A)}{\underline{\qquad\quad}}$$

B. Current Month Virgin Uranium Through the Dissolver Solution Tank
  (1) Gross uranium removed
  (2) Uranium analytical bias as a % recovery
  (3) Recycle

$$\underset{(1)}{(\underline{\qquad})} \times \underset{(2)}{\left(\frac{100}{\underline{\qquad}}\right)} - \underset{(3)}{(\underline{\qquad})} = \underset{(B)}{\underline{\qquad\quad}}$$

C. Fiscal Year to Date Cumulative Pu/U Ratio
  (1) (A) Virgin plutonium removed in current month
  (2) (B) Virgin uranium removed in current month
  (3) Virgin plutonium removed in previous months of this fiscal year (C 1 & 3 on the previous month's worksheet.)
  (4) Virgin uranium removed in previous months of this fiscal year. (C 2 & 4 on the previous month's worksheet.)

$$\frac{\overset{(1)}{(\qquad)} \quad \overset{(3)}{(\qquad)}}{\underset{(2)}{(\qquad)} \quad \underset{(4)}{(\qquad)}} = \underset{(C)}{\underline{\qquad\quad}} \text{ grams/pound}$$

D. Plutonium Charged in Current Month
  (1) Total uranium charged in this fiscal year, including the current month. (D 1 from last month work sheet plus current month.)
  (2) FY to date ratio (C above)
  (3) Ratio calculated plutonium charges for the Fiscal Year, exclusive of current month (D 3 & D from last worksheet).

$$\underset{(1)}{(\underline{\qquad})} \times \underset{(2)}{(\underline{\qquad})} - \underset{(3)}{(\underline{\qquad})} = \underset{(D)}{\underline{\qquad\quad}} \text{ grams charged in current month}$$

Figure 2

RATIO METHOD
PLUTONIUM CHARGED TO DISSOLVERS
SIMPLIFIED WORK SHEET FOR 100 PER CENT WEIGHTED APPLICATION OF ISOTOPE CORRECTION FACTOR

A. Isotope Correction Factor FY to date
   (1) Gross plutonium product loaded out FY to date
   (2) Gross plutonium count (ATTC) FY to date
   (3) Plutonium analytical bias as a % recovery
   (4) Radioassay bias as a % recovery

$$\frac{\overset{(1)}{(\qquad\qquad)}}{\underset{(2)}{(\qquad\qquad)}} \times \frac{\overset{(4)}{(\qquad\qquad)}}{\underset{(3)}{(\qquad\qquad)}} = \underset{(A)}{\underline{\qquad\qquad}} \text{ ICF FY to date}$$

B. Virgin Plutonium Through Dissolver Solution FY to date
   (1) Gross plutonium count (ATTC) through dissolver solution FY to date
   (2) ATTC analytical bias as a % recovery
   (3) (A) Above
   (4) Recycle plutonium FY to date

$$\frac{\overset{(1)}{(\qquad\qquad)}}{\underset{(2)}{(\qquad\qquad)}} \times 100 \times \underline{\overset{(3)}{(\qquad\qquad)}} - \underline{\overset{(4)}{(\qquad\qquad)}} = \underset{(B)}{\underline{\qquad\qquad}} \text{ gms}$$

C. Virgin Uranium Through Dissolver Solution FY to date
   (1) Gross uranium FY to date
   (2) Uranium analytical bias as a % recovery
   (3) Recycle uranium FY to date

$$\frac{\overset{(1)}{(\qquad\qquad)}}{\underset{(2)}{(\qquad\qquad)}} \times 100 - \underline{\overset{(3)}{(\qquad\qquad)}} = \underset{(C)}{\underline{\qquad\qquad}} \text{ pounds}$$

D. FY To Date Pu/U Ratio

$$\frac{\overset{(B)}{(\qquad\qquad)}}{\underset{(C)}{(\qquad\qquad)}} = \underset{(D)}{\underline{\qquad\qquad}} \text{ g/\#}$$

E. Plutonium Charged FY to date
   (1) Pile stated uranium FY to date
   (2) (D) above

$$\frac{(1)}{(\qquad\qquad)} \times \frac{(2)}{(\qquad\qquad)} = \underset{(E)}{\underline{\qquad\qquad}} \text{ grams}$$

F. Plutonium Charged in Current Month
   Current Month. (E) – Previous month. E = ________ grams

Figure 3

error in the gross uranium and plutonium figures are identical and since they are in both the numerator and the denominator in step C cancel out. This is the easiest way of properly weighting each set of analytical results for throughput represented.

The virgin plutonium, A, passing through the dissolver solution tank is calculated by correcting the gross plutonium for analytical bias and for any material included other than virgin feed. The analytical includes several corrections: (1) americium-curium, (2) bias measured in radioassay method, (3) bias measured in determining isotope correction factor.

The virgin uranium, B, is calculated in similar fashion correcting the month's throughput for analytical bias and non-virgin material.

The ratio for the month is (A) virgin plutonium divided by (B) virgin uranium. In the first month of fiscal year the ratio (C) will equal (A)/(B). And going to the last step, the by ratio quantity of plutonium charged for the first month (D) will equal uranium charged in the month times the ratio (C). In the following months a cumulative type of calculation is made to minimize the effect of inventory lag between charge point and dissolver solution measurement tank. As shown in Fig. 2, after the first month (C) the ratio is calculated on a cumulative basis. In the last month of the fiscal year it will be the ratio for all throughput. Plutonium charged (D) for each month is the cumulative pounds charged times cumulative ratio minus the cumulative plutonium by ratio shown for the previous months.

In reviewing Fig. 2, the potential causes of error can be easily spotted. They are:

1. Plutonium analytical.
2. Uranium analytical.
3. Plutonium recycle.
4. Uranium recycle.
5. Inventory lag between charge point and ratio determination point.
6. Uranium charge reliability.

1. Plutonium Analytical

This determination consists of several phases: Radioassay, americium-curium correction to radioassay, and isotope correction factor for converting counts to grams.

a. *Radioassay.* This method has been closely studied in the last year and a half. It is believed bias can be controlled at a low level and what bias is present can be measured and corrected for. The major potential for bias was thought to lay in the effects of beta present. Recent studies made of standards spiked with varying amounts of beta emitters show the effect to be insignificant at normal concentration levels. Bias in the radioassay method is measured by running standards concurrently during the month to permit month end bias adjustment.

b. *Americium-curium.* The Am-Cm correction has recently been based on the results of a monthly composite analysis. The normal drawback of compositing, namely concentration effect, causes no trouble since only a ratio of Am-Cm counts/min to Pu counts/min is needed for the correction. It is believed that the close control possible on this monthly composite, which includes multiple analyses and concurrent standard analysis for detection and correction of bias, provides a reliable bias for this correction.

c. *Isotope correction factor.* The ICF is determined in the product loadout measurement. It is the ratio of grams to counts/min at that point. The average ICF for the month is used to adjust the gross plutonium, A, Fig. 2, which was calculated during the month based on the previous month's average. The ICF is corrected for any bias measured in the X-ray and radioassay methods used during the month for ICF determination.

There is a lag here between point of application and point of use which does contribute some uncertainty to the monthly calculation. However, on the longer term what error is contributed is automatically adjusted for by subsequent loadout of the material in the lag inventory and its proportionate effect on the month's ICF. Here again there is an affect only when the ICF average for the month is grossly different from the ICF in the lag inventory, and any error will be automatically corrected for in the following month.

2. Uranium Analytical

Uranium has been measured by the density method. Early work done to verify this method included comparative analyses of process samples by both X-ray photometer and new direct coulometer methods. Good agreement was shown. Recent changes in type material and processing conditions in one plant has necessitated changeover to an extraction-X-ray method of analysis to avoid bias due to impurities present. Bias in both methods are measured by running standards concurrently, as for plutonium, to permit month end bias adjustment.

3. Plutonium Recycle

In some months there will be recycle either from the Purex extraction area or from the metal finishing plant that will enter at a point between dissolvers and dissolver solution measurement tank. Such material enters at a measured value. These quantities must be considered in determining a virgin metal ratio as shown in Fig. 2. The quantity of such recycle is becoming less and less (currently less than 0.5 per cent of throughput). At the 0.5 per cent level even relatively large errors in the measurement of these streams have little or no effect on the ratio measurement.

4. Uranium Recycle

A small uranium recycle is always present in recovered acid used for dissolving. Extraction recycle occurs in some months too. All recycle is on a measured basis as with plutonium. Uranium recycle also is less than 0.5 per cent of charges so even fairly large errors in recycle measurement have small effects on the ratio measurement.

5. Inventory Lag Between Charge Point and Ratio Determination Point

This lag does contribute to the monthly calculation but the cumulative basis for calculation is used to prevent any carry over effect in periods over a month. Monthly error contributed is significant only when a gross difference exists between throughput ratio and inventory ratio.

6. Uranium Charge Reliability

Uranium is charged into the piles at an accurately weighted value which is corrected for impurities. Shipments to the separations plant are adjusted for burnout. Charges are believed accurate to ±0.1 per cent which is the accuracy of scales used for receipt measurement at HAPO.

FUTURE IMPROVEMENT IN PLUTONIUM METHOD

It is hoped that the radioassay method can soon be replaced with a more accurate direct method such as the coulometer. With the radioassay method dependent upon final product data for the counts-to-grams conversion factor (ICF), short term accuracy will always be sacrificed. There is also in this plutonium method the problem of weighted application of the (ICF) numbers. This weighted application is important for improper weighting precaution can result not only in short term inaccuracies but long term uncertainties as well. Weighted application of ICF means giving all ICF analyses the mathematical weight proportionate to the quantity of material on which it was determined; and likewise in using the product loadout weighted ICF in the dissolver solution measurement on a quantity of material equal to the quantity on which that conversion factor (ICF) was determined.

In the present system used at HAPO (Fig. 2), constant batch size and constant throughput are depended upon to give the proper weighted ICF application. This method has been verified by a 100 per cent weighted system as shown in Fig. 3. The two procedures agree well on the long term but showed as much as one per cent difference in one month in which throughput was not normal. It is intended to go to the 100 per cent weighted system shown in Fig. 3 at the start of the next fiscal year. Of course, any time that a coulometer measurement is possible for routine usage, the radioassay-ICF system would probably be dropped.

In Fig. 3, it can be seen that all parts of the ratio method calculation are done in a cumulative fashion for the fiscal year. In A, the ICF is calculated by dividing cumulative grams by cumulative counts, both appropriately corrected for measured bias. In B, the cumulative plutonium count as measured at the dissolver solution tank is converted to virgin plutonium by use of the ICF from A and appropriate corrections for measured bias and recycle. In C, the virgin uranium is calculated as in the system in use (Fig. 2) but for the FY to date. The FY to date ratio is calculated in D, and in E and F the plutonium charged for the FY and for the current month are calculated.

SUMMARY

A good separations plant receipt measurement is required. The development of such a control must be applicable to the facilities provided. These facilities did present limitations associated with volume measuring, sampling, and analysis. The summation of these uncertainties left much to be desired. The dominating characteristic of the input measurement on control necessitated resolution of the problem.

Two approaches were feasible as a means of solution: (1) reduction of error in the three phases of the measurement method, and (2) elimination of some of the phases from consideration. Several years of effort clearly indicated that error reduction potential was limited to analytical. The ratio method which has been adopted utilizes both approaches in that analytical methods have been improved, and sampling and volume effects have been reduced to a minimum. The ratio method is therefore properly classed as a "design around a problem" in which objectionable limiting factors are eliminated from the calculations.

We believe the ratio method provides a specific improved separations receipt measurement. We also consider that the basic approach is capable of adoption to other problems and offer it to you for such application as you may find of benefit.

DETERMINATION OF A FACTOR ASSAY FOR URANIUM REGULI

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Abstract

The reduction of uranium tetrafluoride by magnesium, in the production of uranium metal, yields an aggregate of varying degrees of purity. The precision of records data is dependent upon the reliability of a nondestructive evaluation technique for routine production. The development of such a technique which has since been applied is described herein. Its importance in materials accountability applications is even more apparent in the subsequent processing steps, wherein the bias of regulus evaluation may well exceed all other measurement errors combined.

Introduction

Until the results of the evaluation described herein became available, it was the practice at this site to assign a single factor assay to the entire production of uranium reguli. This factor had not been substantiated by measurement data for some period of time. Because the use of an inaccurate factor might cause anomalies in both the SS materials accountability program and the interpretation of quality control data, the Accountability Department, with the concurrence of the Oak Ridge Nuclear Materials Management Office, devised and executed a program for the determination of a reliable factor.

The program included the grading of reguli on the basis of external slag, the sampling and subsequent dissolution and assay of representative reguli, and the casting of all-reguli ingots, with the assay of the charge being calculated from the ingot weight and the weight and assay of the residue. The assay determined by dissolution was compared with the assay determined by casting.

Reguli (or derbies) produced in slag-lined steel retorts were graded for exterior slag prior to consumption in the recasting operation. The grades of "good," "fair," and "poor" were established on the basis of appearance. Derbies in the "poor" category are now generally subjected to a shock treatment operation for the removal of excess slag. They are then reclassified into the "fair" category for all future considerations. The derbies used in this study were not shock-treated, however.

Objectives of Work Completed This Quarter

To obtain an average derby uranium assay for each quality grade of derbies produced.

Summary of Results Obtained This Quarter

It is concluded that the uranium assays of the three Metal Production Plant grades of derbies are different and that one average number cannot be permanently applied to any grade. A general factor can, however, be applied to the over-all production, based on the production ratio of each grade for the given time period.

The average assays for the Metal Production Plant derby grades are as follows:

1. Good	99.592% uranium
2. Fair	99.189% uranium
3. Poor	97.402% uranium

It is believed that the Book-Physical Inventory Difference in the recasting area has been reduced because the new assays more nearly represent the actual uranium content of the material consumed in the recasting area.

Pilot Plant and Laboratory Analyses and Tests

A total of 23 derbies from routine Metal Production Plant production were selected from the derbies in two Production Orders to serve as the basis of the evaluations. These derbies were equally distributed among the three derby grades (Fig. 1) and should provide sufficient coverage for the average assay in each quality grade.

FIGURE 1 Derbies Arranged by Quality Grades. The Grading Criterion was the Amount of Exterior Slag on a Derby.

A. Evaluation by Dissolution

Sectors of six derbies were cut in the vertical plane by a power hacksaw. Each piece represented approximately one-twelfth of a derby in net weight and was selected to represent the particular derby quality. The derby sections were weighed on an electronic scale to the nearest 1/100 gram and were individually dissolved in nitric acid. Heat supplied through a stainless steel steam coil and small amounts of phosphoric acid were used to aid in the dissolution. The volume of solution was determined from the specific gravity, and the weight (to the nearest 1/100 pound) was obtained on a Sampling Plant scale. Six samples were prepared to represent each solution - two for specific gravity determination (to five significant figures) and four for volumetric determination of uranium. The weight of solution, the weight of metal dissolved, the specific gravity, and the concentration of uranium in the solution were used in determining the assay for the derby.

As an aid in reducing the dissolution time for the derby pieces, each pie-shaped section was subdivided by a horizontal cut. The resulting top portion of the derby section represented about one third of the original sector weight. The bottom portions of these derby sections were at first assumed to be pure metal, but were later determined to be of a lower assay.

The final results of the derby assay obtained after dissolution are listed in Table 1.

B. Evaluation of Slag Obtained During Dissolution

When the results of the first derby dissolution became available, it was apparent that the uranium content determined was lower than the anticipated factor by an amount far in excess of the expected limit of error. An additional method of evaluating the maximum uranium content was considered necessary.

A large quantity of undissolved magnesium fluoride sludge was isolated from one of the solutions of poor grade derbies. Thus, an alternative *approximate method of evaluating the derby assay* became available. This method, supplementary to the regular dissolution evaluation, is based upon an assay of the uranium in the insoluble

TABLE 1 Evaluation of Metal Production Plant Derbies by Dissolution
(Results are in weight per cent on a uranium basis)

Derby Grade	Derby No.	Evaluation No.	Derby Assay	Grade Avg
Good	H-7951	3	99.779*,†	99.6105
	H-7870	4	99.442*	
Fair	H-7874	5	99.322*,†	99.3765
	H 7929	6	99.431*	
Poor	H-7857	7	96.143‡	97.1835
	H-7944	8	98.224‡	

* Bottom assumed to be 100 per cent or pure metal.

† Verification of bottom assay invalidated due to evaporation of samples.

‡ Bottom assay evaluated and included.

TABLE 2 Dissolution Evaluation vs Sludge Evaluation of Metal Production Plant Derbies
(Results are in weight per cent on a uranium basis)

Derby Grade	Derby No.	Evaluation No	Dissolution Assay	Max U Content Based on Sludge Assay
Fair	H-7874	5	99.322*,†	99.470
	H-7929	6	99.431*	99.609
Poor	H 7857	7	96.143‡	98.725
	H-7944	8	98.224‡	98.298

* Bottom assumed to be 100 per cent or pure metal.

† Verification of bottom assay invalidated due to evaporation of samples.

‡ Bottom assay evaluated and included.

sludge. The method assumed the upper limit of uranium content to be equivalent to the total derby section weight, less the dry insoluble magnesium fluoride weight. A comparison of these data with the dissolution value is given in Table 2.

The differences between the uranium assay and the theoretical maximum uranium content ranged from 0.074 to 0.582 per cent. Since in the maximum uranium column all of the weight except the sludge was assumed to be solid uranium metal, the discrepancies between the two sets of values probably resulted from (1) slag that dissolved during dissolution and (2) uranium that had been in the form of U_3O_8.

C. Evaluation by Recasting

Six all-derby ingots were cast in the Pilot Plant to provide a second basic method of evaluating the uranium content of the Metal Production Plant derbies. The crucible charges were composed of four Metal Production Plant good or fair quality derbies, or three and one-half poor quality derbies for the respective ingots. Duplicate poor derby charges were successfully cast, although the quantity of slag present in the derby required a reduction in the remeltable weight. The finished ingots produced required only normal cropping and were acceptable for rolling.

The crucible burnout, floor sweepings, and furnace lid sweepings were combined, blended, weighed to the nearest 1/10 pound, and analyzed volumetrically for uranium content. The total net weight of the residues produced in the remelting operations, exclusive of ingot cropping, varied from 23.5 pounds to 149 pounds net per ingot and reflected the derby quality.

The assays of derby grades as determined in the recasting evaluation, are listed in Table 3.

Discussion

The final assay value assigned to each derby grade was determined from both the dissolution and the recasting evaluations. The number of derbies represented by each evaluation method was considered important to the final value, since a large range or spread of uranium content exists within a given derby grade. The assumption that the bottom section of the good and fair derbies involved in the dissolution evaluation consisted of pure uranium was considered to have contributed to a high assay bias for this method in the respective derby grades. Since four derbies were used in recast and only one in dissolution, the ratio of derbies represented by each type of evaluation was used to reduce the significance of this bias in computing the accepted assay.

The data from each evaluation and the final accepted assay are listed in Table 4.

The assays obtained by the dissolution techniques for the good and fair derbies were slightly higher than those obtained by the recasting techniques, possibly the result of the assumption that derby bottom portions contained 100 per cent uranium. The difference is not considered signifi-

TABLE 3 Evaluation of Metal Production Plant Derbies by Recasting

(Results are in weight per cent on a uranium basis)

Derby Grade	Ingot Number	Derby Assay*	Grade Average
Good	32529	99.617	99.587
	32526	99.557	
Fair	32530	99.482	99.142
	32527	98.802	
Poor	32534	98.530	97.456
	32536	96.382	

* Based on the weight of the cropped ingot (assumed to be 100 per cent uranium) and the weight and assay of the crop and residue.

TABLE 4 Final Evaluation Data of Metal Production Plant Derbies

(Results are in weight per cent on a uranium basis)

Derby Grade	Dissolution Average	Recasting Average	Accepted Assay
Good	99.610	99.587	99.592
Fair	99.376	99.142	99.189
Poor	97.184	97.456	97.402

cant because of the large errors inherent in grading and the large range of assays within each grade.

The values, for the assays of the respective derby grades, are applied to the accountability material balance reports and are not used in computing the derby or ingot yields. The "poor" derby assay is applied to the respective derby weights prior to shock treatment for the purpose of computing derby uranium production. When the derby has been shock treated and weighed, the "fair" derby assay is applied to the new weight for uranium consumption in the recasting of ingots. The difference in uranium content, before and after shock treatment, is charged to the resulting shock treatment sludge.

Future Work

1. To evaluate the residue from the shock treatment of poor quality derbies, through the recovery and subsequent sampling of this residue.
2. To apply the results of the residue recovery to the assay of the derbies so treated.
3. To determine if the uranium assay of poor quality derbies is a function of the ratio between the poor quality derbies produced and the total derbies produced.

DEVELOPMENT OF PRESENT MATERIAL CONTROL ORGANIZATION AT MALLINCKRODT CHEMICAL WORKS

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MATERIAL CONTROL ORGANIZATION

Mallinckrodt Chemical Works has participated in the refining of uranium ores and concentrates since 1942. During this period, several changes in procedures and organizational responsibilities for the management of nuclear materials have been made.

These changes were made for one or two reasons--those necessary as a result of changes and growth in the processing facilities; and more significant, those directed at consolidation of responsibilities and simplification of recording and reporting methods.

The initial process in 1942 consisted of a facility for refining impure black oxide into purified UO_2. During this period, the accountability of nuclear materials was the responsibility of the Plant Superintendent and clerical personnel reporting to him.

In 1943, the activities were expanded to include hydrofluorination, reduction, and casting; and in 1946, the refining activities were moved to a new facility having greater capacity, flexibility, and efficiency in operation. The expansion of facilities made it necessary for the production superintendent to devote more of his time to production functions. It thus became necessary to relieve him of the responsibility for accountability of nuclear materials. This responsibility was transferred to the Cost Accounting Department. The responsibility for production records and production reporting was assigned to a Production Records Supervisor reporting to the Plant Superintendent.

In 1950 and 1951 new facilities for hydrofluorination, reduction and casting were placed in operation. In addition to having greater capacity, the new facilities were more complex and presented problems not yet experienced in material control. It became evident that the Cost Accounting Department did not have sufficient knowledge of processing operations nor sufficient technical assistance to maintain effective control over nuclear materials. At this time, a separate staff group was established to maintain accountability of nuclear materials. The responsibility of production reports and processing records remained with the Production Records Supervisor reporting to the Plant Superintendent. The Supervisor of the staff group in conjunction with the Production Records Supervisor developed recording and reporting procedures to fulfill the AEC requirements for material control. Every effort was made to minimize duplication of records. Despite these efforts some duplication was necessary principally because of the need for records within plant areas.

As the scope of the operations increased during the 1950 to 1954 period and as additional responsibilities were added to the staff group, it became apparent that consolidation of responsibilities for material control and production records and reports would lead to simplification and uniformity of records, elimination of all duplication, and improvements in material control and production scheduling.

With the startup of the Weldon Spring facilities in 1955, the responsibility for material control and all production records and reporting functions were combined under one supervisor reporting to the Production Control Manager. From previous experience it was recognized that the most effective method of obtaining accurate and complete data for material and production control was to have responsible personnel located within the operating units with a centralized control center for overall control, procedures, and consolidation of reports.

The present Material Control Organization at Weldon Spring includes a Supervisor, an Assistant Supervisor, five Chief Plant Clerks, and 13 clerks. The Chief Plant Clerks are located in each of the plant areas. Clerical personnel are assigned to the Chief Plant Clerks and the Control Center as required, dependent on the scope and amount of activity within the areas.

The Weldon Spring site consists of a Feed Preparation and Sampling Plant, a Refinery, a Hydrofluorination Plant, a Metal Plant, two Pilot Plants, a Residue Storage Pit, and Laboratory facilities.

In total, there are eight plants or areas designated as material balance areas; in each, a separate material balance is maintained.

The Feed Preparation and Sampling Plant samples and weighs certain concentrates for payment purposes and prepares and packages concentrates into hoppers for refinery use. Such a plant did not exist in the Destrehan site. Approximately 400 containers are processed daily through this plant.

The refinery digests the concentrates in nitric acid and purifies the solution by extracting into Tributyl-Phosphate-Hexane solution and re-extraction into water. The resultant pure aqueous solution is then denitrated to uranium trioxide. The Weldon Spring refinery has three times the capacity of the Destrehan refinery. The use of hoppers for packaging feed material and product in the Weldon Spring refinery versus the use of drums for the same purpose at the Destrehan site has reduced the number of containers to handle and account-for by a factor of twenty-five for comparable weights.

The Hydrofluorination Plant reduces UO_3 to UO_2 with hydrogen in a fluid bed which is then fluorinated with Hydrofluoric Acid to UF_4. As in the Refinery, the production rate is three times that of the similar operation at the Destrehan site. The use of hoppers rather than drums has reduced the number of feed containers from twenty-five to one and the number of product containers from twenty to one for comparable weights.

The Metal Plant reduces the UF_4 to massive metal which is then scalped to remove surface impurities and imperfections. The clean metal is then extruded into bars for shipment offsite. This plant differs from the Destrehan Metal Plant by eliminating the recasting operation of the metal. Here again, the increase in the size of the unit has decreased the number of units by a factor of ten.

The two Pilot Plants test new processes for further improvements in the operations. The laboratory facilities include an Analytical and a Development Laboratory.

For each of the production and technical plants, or units, a recording and reporting system is required by the AEC, Management, Production Control, and Cost Accounting.

The AEC requires a record and reporting system which will provide current and accurate information as to the disposition and availability of nuclear materials. Monthly material balance reports are required for each operating unit and for the overall Weldon Spring site.

Other AEC requirements are:

1. A monthly report of plant yields,
2. A weekly and monthly narrative report of operations,
3. A daily report on production activities, and
4. Reports of a non-routine nature on a specific subject on relatively short notice.

Mallinckrodt management essentially requires the same records and reports as specified by the AEC and in addition requires reports on Processing Costs, and other data pertinent to the effective and efficient operation of the plants.

Production Control requirements are:

1. To maintain a current comparison of actual versus scheduled: production, shipments, and receipts,
2. Development of operating rates and inventory levels for all production plants,
3. Initiation of Production Orders and the responsibility for the completion of such orders, and
4. Schedule feed material receipts and product shipments.

The responsibilities of the Material Control Organization have been expanded to include completion of the Inventory and Manufacturing Statements exclusive of the dollar values. These data for cost accounting purposes are identical to the values reported in the material balances.

AEC-101 forms must include identification of material transferred so that costs can be properly applied and transferred.

Other requirements of a production records organization are to:

1. Schedule for purchase and delivery dates all processing chemicals, supplies, and reagents used throughout the Weldon Spring site and to maintain adequate inventories of these materials to assure continuity of operations.
2. Maintain, follow-up, and report on costs incurred for other than routine production schedule requirements.
3. Maintain files of all maintenance orders.
4. Accumulate data and perform statistical evaluation of such data.
5. Maintain records for allocation of overtime to production operators.
6. Provide services to operating supervision, such as typing, filing, summarization of data, production on-time determination, and yields of specific phases in the operation.

Both the Destrehan and the Weldon Spring organizations accomplished the requirements of the AEC, Management, Production Control, Cost and Operating Supervision. However, by combining all these responsibilities into a single organization the scope of personal responsibilities has been considerably increased. As much as is possible and practical the responsibilities of the Chief Plant Clerks have been widened to include every phase of the requirements made of the organization.

In the Destrehan organization the plant clerical personnel did not prepare material balance reports. At Weldon Spring each Chief Clerk is responsible for all nuclear materials received, shipped, and on inventory and for the preparation of the material balance reports of his plant. The audit and consolidation of reports is done by the Control Center.

Daily production reports are prepared by the Chief Plant Clerks from accountability records at Weldon Spring in contrast to subsidiary records maintained by plant clerical personnel at Destrehan.

Much of the data accumulated and reported and other functions performed by technical personnel at Destrehan is now being accomplished by the Chief Plant Clerks and the Control Center. Narrative reports of plant operations are submitted by the Chief Plant Clerks to the Control Center for editing and consolidation into required AEC reports. Calculations of equipment "on time" and reasons for lost time are reported by Chief Plant Clerks rather than by technical personnel as was done at Destrehan.

The development of production rates and optimum inventory levels was a management and production supervision function at Destrehan. At Weldon Spring, the Chief Plant Clerks are responsible for these determinations within the framework of established policy.

The reports of official weighing, sampling, and analysis were very limited at the Destrehan site. At Weldon Spring this function has increased to an eight-hour day function.

The Destrehan plant clerical personnel were members of the union and were assigned to specific plants. Cross training was very limited because of union restrictions. All clerical personnel in the Weldon Spring organization are on salary payroll and are cross trained in all functions without restriction.

Meetings are held at least once per week with all Chief Clerks and Control Center personnel for the purpose of discussing new developments and procedures. To effectively perform his function, each Chief Clerk must be fully informed on the developments in the other plants including the one he is primarily responsible for. This exchange of information takes place during the weekly meetings. Training programs and discussions on new procedures and changes in existing procedures take place during the meetings. Many ideas for reducing operating and clerical costs are also generated.

The Destrehan organization included thirteen clerical, ten accountability, and four other personnel for miscellaneous functions of which twenty-one were male and six were female employees.

The combined Weldon Spring organization consists of a total of twenty personnel of which fourteen are male and six are female employees with a reduction of seven male employees over the Destrehan organization. Because of the better quality of personnel in the Weldon Spring organization and a constant critical examination of the methods employed to fulfill the necessary requirements, it was possible to reduce the number of personnel by approximately 30% and at the same time to increase considerably the scope of the organization.

The savings in personnel and increase in scope of responsibilities is in part due to the use of punch card techniques for data recording and reporting. These techniques are being expanded to include all areas of recording and reporting functions.

In summary, the consolidation of responsibilities into a single organization has:

1. Improved communications with the AEC, Management, other contractors, and operating supervisions,
2. Provided quicker solutions to problems by improvement in the calibre and training of personnel, and
3. Establish uniformity in records and reporting procedures.

From our Destrehan experience we have always contended that the responsibilities for the management of nuclear materials can be better fulfilled by personnel who are thoroughly familiar with every phase of the operation. The responsibilities for management of nuclear materials are so closely related to and a part of all operating and reporting functions that separate organizations would of a necessity require duplication in records and efforts.

Mr. John Nicholson will now discuss some of the punch card techniques applied to operating and nuclear material control data.

DATA PROCESSING

During the past few years, the Mallinckrodt Materials Accountability Group has been striving for more complete coordination and streamlining of data collection, recording, and reporting methods. One approach has been the use of IBM punch card methods for recording and handling data to achieve reduction in data handling and to result in a more generally coordinated effort of the group as a whole. The basic problems applied to the IBM approach vary with the system and can in no way be entirely stated by the standard phase "large volume of data". The areas considered for application of IBM data handling methods are characterized by:

1. A small volume of data with highly repetitive use,

2. A small to moderate volume of data with a highly complex data handling and reporting program,

3. A moderate volume of data requiring great detail, or

4. Moderate volume of data where coordination is hampered by geographical location.

Desired characteristics of the systems were that they would achieve a higher degree of report uniformity than the manual systems in use and thus allow for simpler cross training of personnel for all of the plant areas. In addition, by reducing all transactions such as consumption, production, and shipments to a basic format, not only could report formats be greatly simplified, but the collection and handling of data could be greatly standardized and thus simplified.

The systems chosen for discussion in this paper are two extreme phases of the program currently in operation. The first is an example of a system concerned primarily with all transactions in only one balance area. The data are recorded on the production line by operating personnel; control of these data is by materials accountability personnel in the production office. Reports prepared by the IBM Group serve both materials accountability and production needs.

The second case is an example of a system concerned with only one type transaction for the entire Weldon Spring station. The data are transferred from a secondary source which has plant records as a data source. In contrast to the first-mentioned system, this system involves data recording and control by the Materials Accountability Group, and IBM-prepared reports specifically to serve the Materials Accountability needs. As further contrast, the first-mentioned system is characterized by a small volume of data requiring highly repetitive data use in a very complex reporting program, while the second case involves a moderate volume of data, some repetitive data use and great detail.

The Weldon Spring dingot plant provides the best example of the IBM data collection method in use on the production line. The dingot process is a batch-wise operation that involves intricate interplays among a number of intermediate products. Application of IBM methods in this plant was based on the complexity of the required program and the highly repetitive use of plant data as for example product weights and corresponding production dates, and not on the volume throughput of the process. The data collection system in use provides an IBM card for direct data recording for each process step for each batch processed. The cards are pre-punched with a sequential batch number and step number, tabletted, and delivered to the plant production office in excess supply. Each booklet is marked with a unique batch number, and contains a card for each step in the process.

This is by no means a novel approach to collection of materials accountability data, similar programs have been used by other AEC contractors for following batch-type processes. The dingot plant cards, however, in addition to handling materials accountability data, provide space for recording process variables. Process variables have been selected in each step of the operation that are considered important characteristics of the operation to aid in analysis of throughput efficiency and/or are considered to have an effect on product quality.

The method of recording data on the cards is routine for all process steps. Simply stated, materials accountability data are handwritten or machine printed for later key punching and verification, process variables where possible are marked for mark sense punching. In certain cases, lack of mark sense fields has made it necessary to hand write process variables for key punching.

Booklets are given to personnel performing the first process step in scheduled amounts, thus providing direct control of batch initiation. Completed cards are delivered to the production office once daily, while the unused portions of booklets move through the plant with the batches to which they correspond. When batch failure occurs at any point in the process, the last used card or that corresponding to the point of failure is marked to show the type failure and is returned to the production office with the unused portion of the booklet.

The completed cards are used in the production office,

1. For maintaining a perpetual plant inventory,
2. As source data for production reports, and
3. As a convenient file of plant data.

The perpetual inventory is shown by the location of a numbered card in a series of card pockets mounted on a board to represent each plant process step. Batch numbered cards originally used for physically separating batch booklets are removed before release of the booklets to the plant. These separator cards are moved from step to step on the perpetual inventory board as completed process cards are returned from the plant. The board:

1. Shows batch inventory levels at each step as an aid to inventory control,
2. Serves as a locator file for finding test batches in the plant, and
3. In combination with physical inventory information, aids in determining if card return is complete.

Cards received each day are counted by step number and the number of batches processed for each step is recorded with the scheduled number. Weights are tallied for each product and the combination of total product weights compared with the schedule and the number of certain of the batch steps completed compared with the schedule are reported daily.

The cards are accumulated in card files with one file tray per step number. The cards are arranged in sequential batch number order; cards not processed by the IBM Group are kept separate from those having been through the IBM program. These files serve as a complete reference to plant operation and are available for use to anyone desiring information.

Once weekly the unprocessed cards are sent to the IBM Group for preparation of the following routine reports:

1. Material Balance Ledgers - A listing is made for each product summarizing material production, consumption, and transfer. One line is devoted to each batch processed through the step showing weight and final disposition at the time of the report. These listings are developed on a monthly schedule commencing with beginning inventory, showing production in sequential batch number order, and at month end, showing the ending inventory.

2. Scrap Production Lists - A list is prepared in relation to each balance ledger showing weight change in the product at the completion of a process step. These lists are prepared so that batch-wise they compare directly with the corresponding balance ledger.

3. Process Variable Lists - Certain process variables and yields are listed routinely in sequential batch number order for use by operating personnel. Variables selected for these weekly listings change from time to time based on the needs of the operating group.

Special requests for process data are answered by one of two methods depending on the extent of reporting required. Short requests or those requiring information for only a small number of batches can be answered by reference to the card files in the production office. More lengthy or complex requests for data are handled by the IBM Group.

The IBM system concerned with SS material transfers best exemplifies the contrasting extreme to the dingot plant program. The SS materials transfer system embraces all balance areas at the Mallinckrodt accountability station, but is concerned only with transfers of SS materials to and from these balance areas. Data for the system are derived from a secondary type media, shipping documents, which have as data source records made on the production line. Materials accountability personnel originate and control the data, and IBM reports are prepared specifically for use by the same group. To provide completeness in the IBM-prepared reports, a system for accumulating and summarizing ending inventory values has been combined with the transfer system. Basis for application of IBM methods in this case is a moderate volume of data requiring a large degree of detail with moderate repetitive use of data.

The material transfers considered by this system may be separated into three basic activities:

1. **External Receipts - Transfers of material from another accountability station or licensee to the Mallinckrodt accountability station,**

2. **Internal Transfers - Transfers of material between balance areas within the Mallinckrodt accountability station, and**

3. **External Shipments - Transfers of material from the Mallinckrodt accountability station to another accountability station or licensee.**

Data source for the external transactions is the AEC-101 form, while for internal transactions it is the Mallinckrodt shipping order. Data for all material transfers are recorded in a uniform sequence to facilitate simplification of the IBM program and uniformity of IBM produced reports.

Data on shipping documents are transferred directly to punched cards eliminating need for duplication of effort by preparation of special key punch source records. The direct key punching of data is accomplished by use of stencil-like overlays designed to expose only those data required for key punching. Receiver information on incoming 101 forms, shipper information on outgoing 101 forms, and shipper information on internal shipping orders are completed with reference to the pattern specified by the appropriate overlays for these types of documents. The data are then ready for punching directly into cards.

Shipping documents in the three categories are accumulated and once weekly are delivered to the IBM Group for key punching. One card is punched for each batch or lot of material transferred, thus more than one card can result from a single shipping document. Since the shipping documents are audited by materials accountability personnel prior to delivery for key punching, and all key punching is machine verified by the IBM Group, only brief checks are made on punched information by the material accountability group. The checks are:

1. Comparison of total weight by type transfer between an adding machine tape and a direct IBM list of cards punched to assure that the proper number of cards have been produced, and

2. A rapid scan of the IBM list in search of gross oversights or misreadings of control information by the key punchers.

Reports are prepared by the IBM Group each month after transactions for the month are completely recorded in punched cards. These reports make up the major portion of the MCW Station Report. Briefly, the listings are:

1. Batch detail and summary by material of external receipts and removals according to the Station Report format defined in Chapter 7402-021A in the AEC Materials Accountability Manual,

2. Composition of Ending Inventory listing, and

3. Summary of Balance Areas, a one line summary by balance areas showing beginning and ending inventories; receipts and removals classified as internal, in area, and out area; production consumption, MUF, and limits of uncertainty. The last four items mentioned are at this time manually typed on the IBM reports.

In addition to the above-mentioned reports, internal transfers are listed according to the format used for external receipts and removals. These internal transfer listings are used by the material accountability group for final manual area balancing.

A new program currently in the experimental stage, permits recording data from the four basic transactions: production, shipment, receipt, and consumption. Data surrounding these four basic transactions will be recorded for all SS materials exclusive of in-process type materials at the Mallinckrodt accountability station. The basis for application of IBM methods in this case is a moderate to large volume of data requiring a moderately complex program with moderately repetitive use of data. The expected savings of this program include:

1. Elimination of internal shipping orders,

2. Complete IBM preparation of required detail listings for all balance areas,

3. Extensive reduction in duplication currently caused by the geographic separation of records and in a few cases, necessary correlations with IBM systems, and

1. The radiation detector must have sufficient sensitivity and accuracy.
2. The natural radiation background must either be negligible or must be corrected for.
3. The counter geometry must be constant.
4. The sample geometry must be constant.
5. The self absorption of the sample must be constant or corrected for.
6. The radioactive element to be assayed must be of constant isotopic composition. This will be true for uranium with a constant enrichment factor.

Both methods described here are based on the measurement of the gamma radiation from U^{235}.

Total Loading Gage

To determine the total loading of fuel fillers the detector must be large enough to absorb essentially all of the gamma radiation coming from the sample. In addition the detector must be equally sensitive to the radiation coming from all portions of the filler, so that some portions of the filler will not influence the reading more than others.

After surveying available gamma radiation detection methods, an Ohmart radiation detection system was selected as the most suitable for this gage.

An Ohmart detector, or cell, converts radioactive energy directly into electric energy. It contains two electrodes which have different work functions separated by a gas which is ionized by radiation. When ionization occurs the positive ions are attracted to the electronegative electrode and the electrons are attracted to the electro-positive electrode, thus generating an electric current. This current is proportional to the intensity of the radiation.

The advantages of the Ohmart system for this application are that the detectors are available in large size, they are rugged, they require simple circuitry, and they have a high stability. These outweigh the disadvantages of a somewhat lower sensitivity than scintillation counters, and that it is a rate type device which is inherently less accurate than a pulse counting system for moderate radiation intensity levels.

The detector assembly, shown in Fig. 1, consists of 2 Ohmart LL-4 assemblies consisting of 4 stacked LI cells. Each assembly is $2\frac{1}{2}$ in. in diameter and 31 in. long. One fourth inch of lead shielding is placed around the cells to protect them from stray radiation. A sliding sample changer provides a means of placing all the test samples in exactly the same location, thus keeping the counting geometry constant. This sample changer also provides a means of changing samples rapidly to prevent the meter from going off scale while there are no samples in the gage.

A schematic diagram of the entire gage is shown in Fig. 2. The output of all the LI cells, connected in parallel, is connected to the input of a Beckman micro-microammeter. Also connected to the input is an Ohmart compensating cell containing a small radium source. This cell has reversed polarity, and in effect generates a negative current to cancel out part of the current generated by the LI cells. This results in zero supression so that a small change in radiation level can be spread over the entire meter range. The amount of current generated by the cell and likewise the amount of zero supression may be varied by varying the location of the radium source.

Radiation intensity readings could be taken directly from the micro-microammeter. However, due to the random nature of the radiation, there will be a random fluctuation in the radiation intensity so that the instantaneous meter reading does not have sufficient accuracy to meet the requirements set for this gage. To increase the accuracy of the gage a Minneapolis Honeywell Brown integrator is used. The input circuit of the integrator, which was designed to operate in conjunction with a Brown ElectroniK recorder, was modified to accept the signal direct from the micro-microammeter. Using the integrator, the radiation intensity is integrated over a 2-min period.

The micro-microammeter is a rate type device with fairly long time constant. Therefore it is necessary to allow the instrument to stabilize after every change of sample before readings are taken. To reduce this time to a minimum and to prevent readings from being taken before the instrument has come to equilibrium, a low level limit switch and a precycle timer are included in the gage.

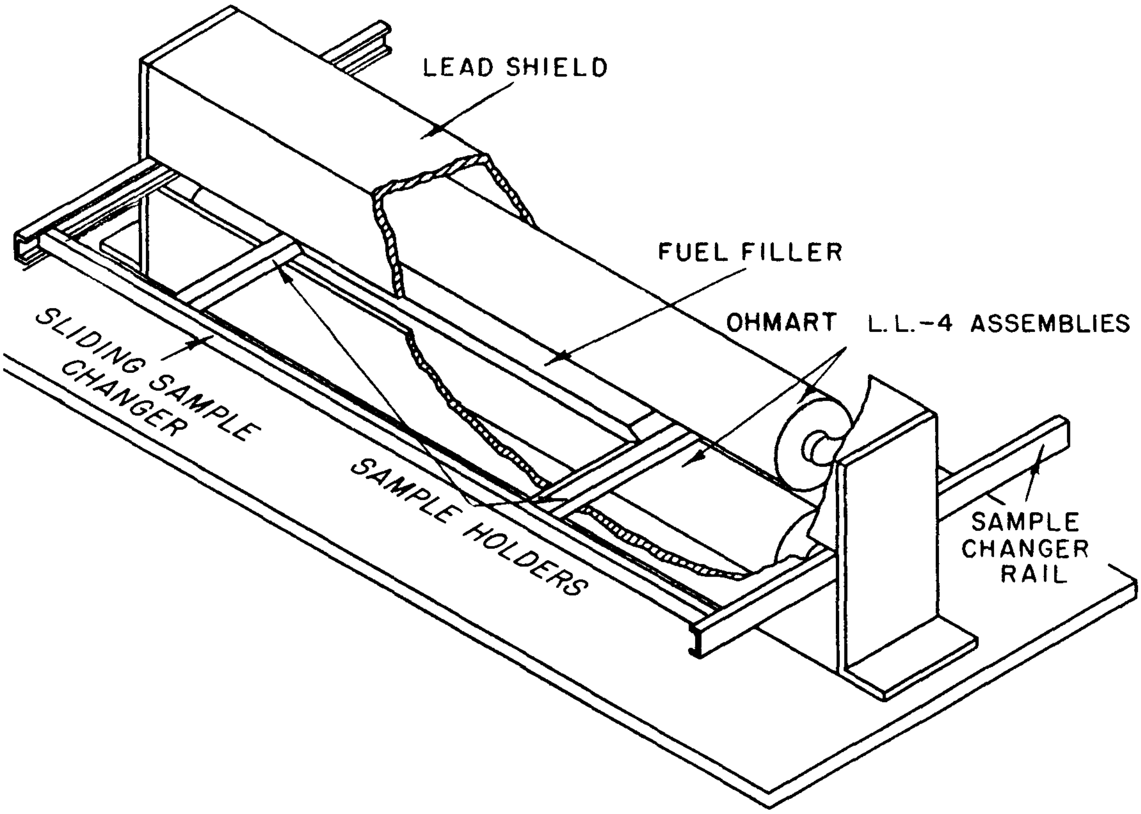

Fig. 1—Detector assembly U^{235} total loading gage.

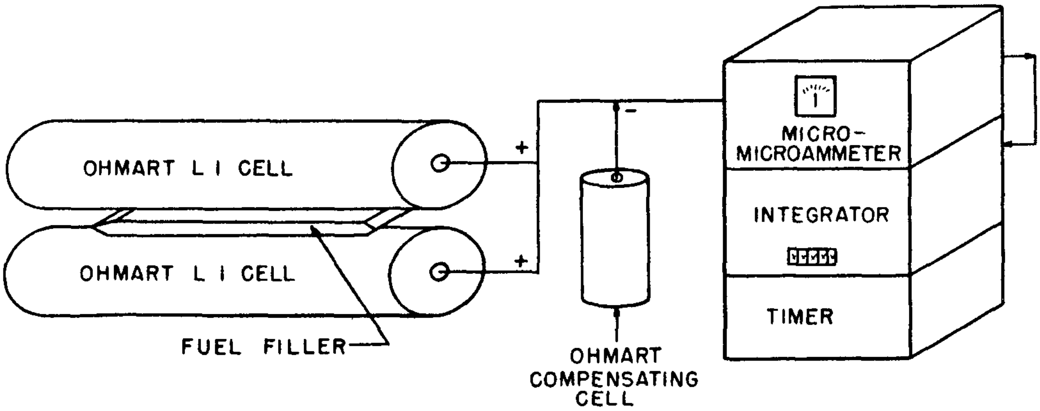

Fig. 2—Uranium-235 total loading gage.

To meet the constant sample geometry requirement set forth in the design criteria, the standards used to calibrate the gage must be the same size and shape as the fillers to be inspected. Any filler which meets the dimensional tolerances set forth on the filler drawings (typical values are length + .000 in. – .005 in., width + .000 in. – .003 in., and thickness ± .002 in.) will have constant geometry as far as the gage is concerned. New calibration standards must be made up for each new size and shape of fillers.

The self absorption of the radiation in the sample is a function of sample thickness. If the fillers to be inspected have a thickness variation of more than .002 in., corrections must be made. This is done by making separate calibration curves for the different thicknesses.

Nine standards are used to calibrate the gage. These standards are the same length and width as the fillers to be inspected and have the following lengths and thicknesses.

Standard	Loading	Thickness
1	107% of nominal	nominal + .003 in.
2	107% of nominal	nominal
3	107% of nominal	nominal – .003 in.
4	nominal	nominal + .003 in.
5	nominal	nominal
6	nominal	nominal – .003 in.
7	93% of nominal	nominal + .003 in.
8	93% of nominal	nominal
9	93% of nominal	nominal – .003 in.

Nominal refers to the nominal value of the fillers to be inspected.

A typical calibration curve is shown in Fig. 3. These curves were made by plotting the loading versus integrator readings for each of the three standard thicknesses. Curves for intermediate thicknesses are interpolated.

Homogeneity Gage

To measure the uranium homogeneity of the fuel fillers, a collimated scintillation counter was chosen because of its high sensitivity. The detector assembly, shown in Fig. 4, consists of two $\frac{1}{2}$ in. diameter by 1 in. thallium activated sodium iodide crystals mounted on DuMont 5692 photomultiplier tubes, and housed in conventional detector housings.

Lead shielding is used to collimate the radiation reaching the detectors in such a way that they will detect only the radiation coming from a $\frac{1}{2}$ in. diameter cylinder through the thickness of the filler. This provides a means of keeping the sample and counter geometry constant except for thickness variations. The lead shielding also helps reduce the counter background by providing shielding against stray radiation.

A schematic diagram of the gage is shown in Fig. 5. One thousand volts are supplied to both detectors from a common high-voltage supply. The preamplifier (cathode follower) serves as an impedance matching device to couple the high impedance of the detector outputs to the low impedance input of the main amplifier, where the signals from the detector are amplified to a level at which they can be counted by the decade scaler. A preset timer is used to control the counting period of the scaler. A 2-min counting period is used. The total count received during this period is proportional to the uranium concentration at that spot in the filler. As with the total loading gage, thickness corrections must also be made. The calibration of this gage is the same as that described for the total loading gage, except no restriction need be placed on the length or width of the standards or samples to be inspected. A typical calibration curve is shown in Fig. 6.

Using the local uranium concentrations as determined by this gage and the weight of the filler, the total uranium loading of the fillers may be calculated in a manner similar to that used with the X-ray fluorescence method. This enables one to determine both the uranium

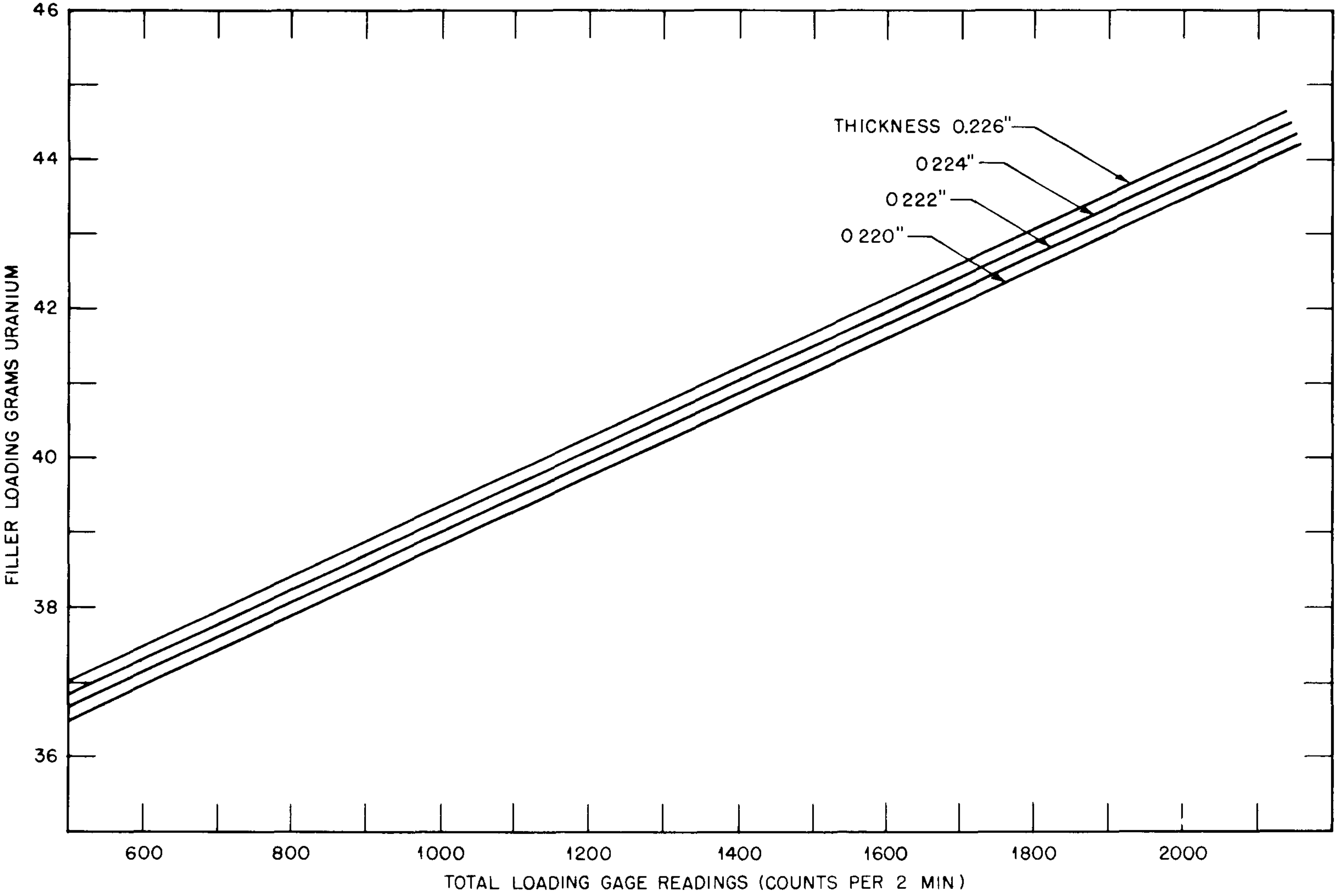

Fig. 3—Typical calibration curve, total loading gage (PWR).

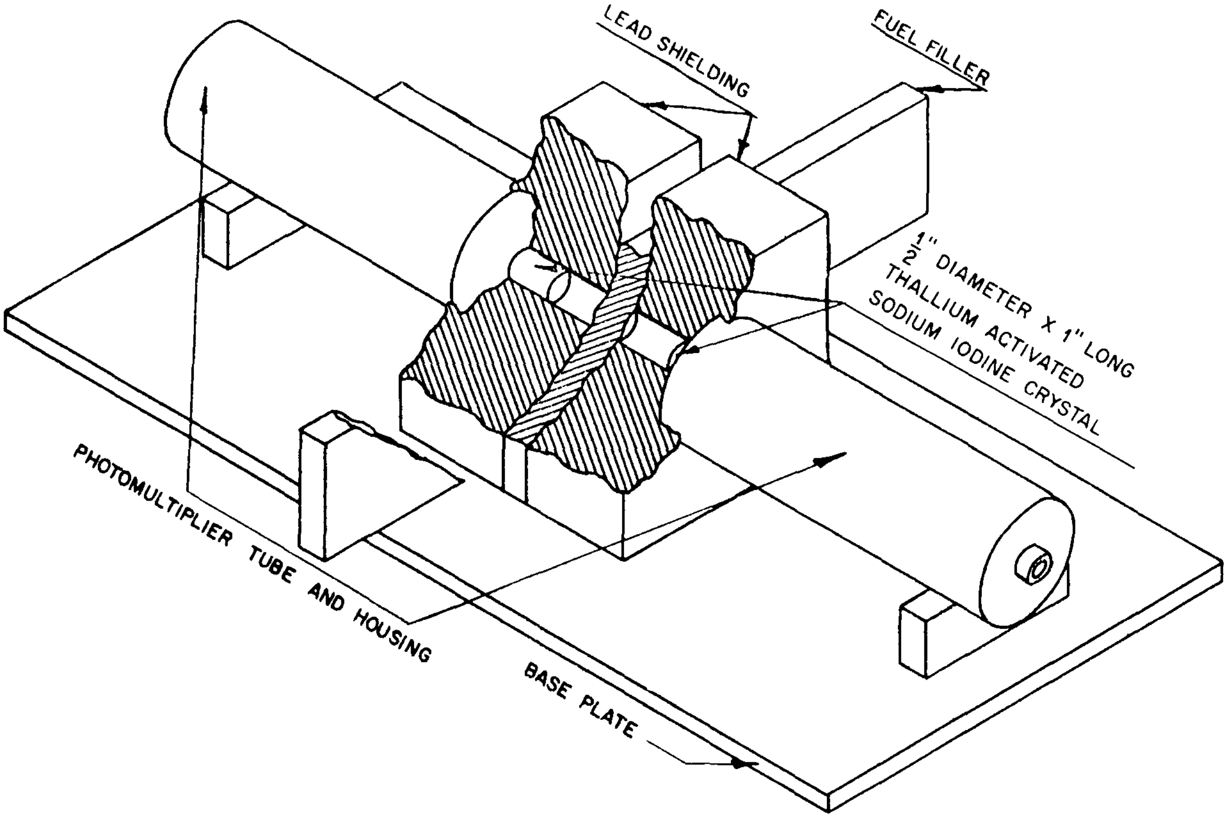

Fig. 4—Detector assembly U^{235} homogeneity gage.

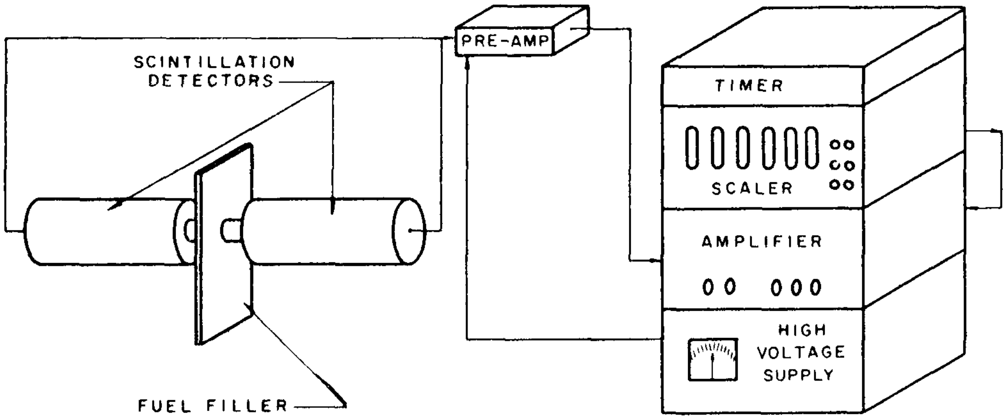

Fig. 5—Uranium-235 homogeneity gage.

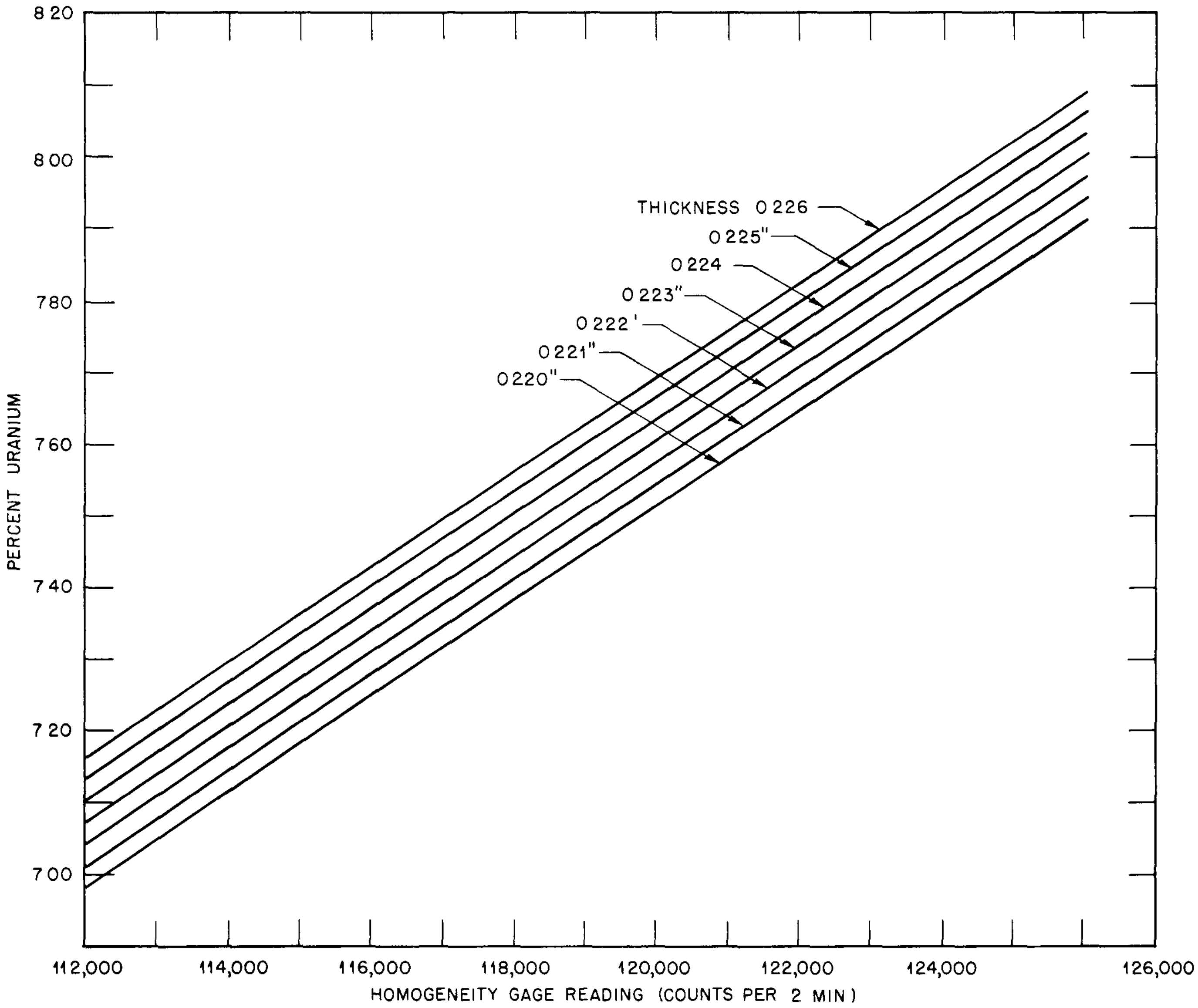

Fig 6—Typical calibration curve, homogeneity gage (PWR)

homogeneity and total loading by the use of only one nondestructive inspection technique. However in this case, the total loading is only a calculated value instead of the actual loading.

EVALUATION

To determine the capabilities of the total loading gage, 250 PWR Core I Seed II production fillers were checked. These fillers are approximately $11\frac{3}{4}$ in. long, 2 in. wide, and .223 in. thick. The loadings of the fillers used as standards were determined by wet chemistry methods (estimated precision ± 0.5% at 2-Sigma). Filler loadings as obtained with the total loading gage were compared with the values calculated from X-ray fluorescence analysis (estimated precision ± 1.2% at 2-Sigma). The estimated precision of the total loading gage was found to be 1.4% at 2-Sigma.

Eighty-five of these fillers were read on the homogeneity gage. Standards were again analyzed by wet chemistry methods. The homogeneity gage readings were compared with X-ray fluorescence readings on an adjacent sample (estimated precision ± 1.0% at 2-Sigma). The estimated precision of the homogeneity gage was found to be ± 1.1% at 2-Sigma.

A cost analysis comparing the two autoradiometric assay gages with X-ray fluorescence is shown in Table I.

Table I. COST ANALYSIS OF ENRICHED URANIUM ASSAY BY AUTORADIOMETRIC METHODS AS COMPARED TO X-RAY FLUORESCENCE*

	Determination of homogeneity only		Determination of total loading only		Determination of both total loading and homogeneity	Savings over X-ray fluorescence	
Method	Cost	Precision	Cost	Precision	Total Cost	Total	%
X-ray fluorescence	$12†	±1.0%	$16.47‡	+1.2%	$16.47‡		
Total loading gage			7.42§	±1.4%			
Total loading and homogeneity gages	2†	±1.1%	7.42§	±1.4%	9.42	$ 7.05	43%
Homogeneity gage	2†	+1.1%	6.47‡	+1.3%	6.47‡	10.00	60%

* The cost figures given are per ingot. Twelve fillers are made from each ingot. Precision figures are at 2-Sigma.

† Based on three samples per ingot.

‡ The total filler loadings are calculated from average ingot uranium concentrations.

§ The total loading of each filler is determined separately.

CONCLUSIONS

A savings of 60 per cent of uranium assay costs can be made by replacing X-ray fluorescence analysis with the homogeneity gage. Using this gage both uranium homogeneity and total loading can be calculated with an accuracy equivalent to that of the X-ray fluorescence method.

The total loading gage provides a method of determining individual filler loadings in cases where, due to gross ingot inhomogeneity or other reasons, filler loadings cannot be accurately calculated from the analysis of a few samples per ingot.

ACKNOWLEDGMENT

Much of the original development work on the total loading gage was performed by R. C. Barry. Much of the experimental work and actual construction of the gages was done by D. R. Agosti. The evaluation of both gages was performed by C. E. Allbaugh.

EVALUATION OF THE PURCHASE METHOD FOR THE SAMPLING AND ANALYZING OF URANIUM CONCENTRATES AT GRAND JUNCTION

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INTRODUCTION

This paper describes the evaluation of the weighing, sampling and analytical procedures used to purchase domestic uranium concentrates. Approximately 275 million dollars worth are purchased per year.

In May of 1948 the American Smelting and Refining Company was selected to operate the uranium concentrate sampling and analytical facilities at Grand Junction, Colorado. In February, 1956, Lucius Pitkin, Inc. took over this operation. By February, 1957, the uranium concentrate receipts had increased considerably and the need for expanded evaluation studies was apparent. A separate Evaluation Group was formed to carry out the expanded studies. The program was aimed at the evaluation of present sampling and analytical methods and to investigate new methods which show promise of increasing accuracy and decreasing costs.

SUMMARY

From the evaluation of the uranium concentrate sampling and analytical methods used at Grand Junction, the following conclusions are drawn.

1. The weighing of the lots of uranium concentrate is accurate, being maintained so by the use of Class C check weights to calibrate scales. The standard deviation of weighing an average lot is less than 2.0 pounds per 10,000 pound net wet lot size.
2. The taking of the primary sample was found to be accurate by comparing the auger sampling of a series of single drums to the sampling of these drums after thoroughly blending the contents of each drum.
3. The precision of the method for taking the primary sample is good as determined by duplicate sampling a series of drums. For the lots tested the standard deviation of the auger method for taking the primary sample was found to be less than 4.2 pounds of U_3O_8 for a 10,000 pound net wet lot size.
4. The official purchase moisture determination and sample preparation procedures were found to be accurate. This was determined by comparing the wet basis assay of the official purchase sample to "pipe" samples taken of the blended lot sample prior to the moisture determination. The assaying of the "pipe" samples eliminated all drying and sample preparation errors for a true basis on which to determine the accuracy of this phase of the "Official Purchase" sampling method.

5. The precision of the lot moisture procedure was found to be good. The standard deviation of the lot moisture procedure obtained from replicate samples was in most cases less than 0.03% H_2O, which is equivalent to less than 0.03% of the U_3O_8 contained in a 10,000 pound net wet lot. The precision did not change significantly at different moisture levels up to 10% H_2O.

6. The analytical procedure for U_3O_8 in these concentrates is accurate, as no significant difference between two distinctly different methods of analysis, the "phosphate precipitation" method and the "zinc reductor" method was found. The latter method of analysis is the "Official purchase" procedure for U_3O_8 at Grand Junction. The precision of the U_3O_8 analytical method is excellent. The standard deviation derived from duplicate determinations carried out on the same day by different chemists (official purchase procedure) was 0.03% U_3O_8 absolute.

   Single determinations with several days intervening yielded a larger standard deviation of 0.09% absolute.

7. The precision of the sampling and U_3O_8 analysis as one overall operation was obtained by performing duplicate measurements of each step on the same days. The standard deviation of this overall operation was found to be 4.94 pounds of U_3O_8 on the basis of a 10,000 pound net wet lot. This standard deviation is equivalent to 0.07% of the pounds of U_3O_8 contained in the lots. Despite the varying chemical and physical characteristics of these uranium concentrates, the precision and accuracy of the overall operation is excellent.

   Additional studies are under way which will cover the evaluation of the overall sampling method for accuracy, and the precision of the overall sampling method on a day by day basis. Also, improvements in the present operations will be tested.

   The quality control program is being continued in the sampling and analytical operations to maintain proper control in all phases of these functions.

URANIUM CONCENTRATE CHARACTERISTICS

Domestic uranium concentrates are complex salts of uranium. The concentrates as received vary from 1/4 inch material down to micron size; the majority are less than 60 mesh in size. The color may vary from black thru brown to grey, red, orange, yellow and tan. The concentrates have an average density of about 82 pounds per cubic foot. The lots of uranium concentrates average about 80% U_3O_8 and the lot moisture may vary from 0.00% H_2O to 9.6% H_2O. Also, these concentrates may contain from 0.1% to 9.0% Na and 0.1% to 13.0% SO_4. Other impurities, which are found in varying smaller amounts, are vanadium, iron, phosphorus, arsenic, molybdenum, calcium, boron, etc.

Papers previously given at this meeting have clearly shown the effect that changes in the water content of ambient air have on the drying of these concentrates. (1) This effect will be covered in this paper only as it relates to the evaluation of our sampling method.

SAMPLING REQUIREMENTS

The physical operation of the purchase program consists of the receiving, sampling, analyzing and shipping of these concentrates.

In the beginning of the present procurement program no extensive industrial experience was available in the actual sampling of these western uranium concentrates. The sampling method chosen had to meet the following requirements:

1. The sampling method must be capable of accurately determining the uranium content of a lot composed of one to many drums.
2. The sampling is to be performed in transit.
3. The concentrates are to be transported in sealed drums due to their high unit value.
4. Air contamination and radiation due to the uranium concentrates must be maintained below maximum permissible limits.
5. The equipment and working areas must be capable of thorough routine cleaning to prevent contamination from lot to lot.

   These requirements must be met as economically as possible.

Other factors which influenced the choice of sampling method are:

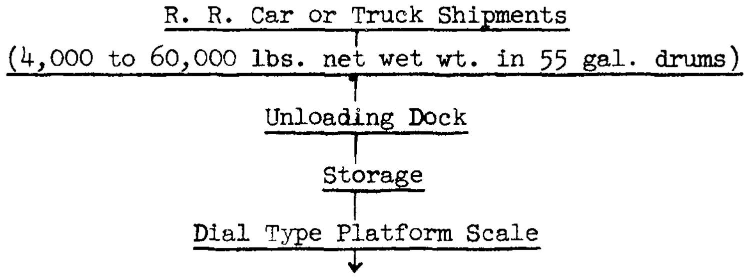
1. Uniformity of % U_3O_8 and % H_2O in the concentrates.
2. Small particle size. Specifications state that all material as received shall pass through a 1/4" mesh sieve. Most of the mills ship a fine product, up to 80% through 100 mesh in several cases.
3. The experience of the non-ferrous metals industry in sampling metal concentrates of various kinds by the auger was also considered. This experience had been gained by years of auger sampling non-ferrous concentrates.

The requirements for the sampling and the characteristics of the uranium concentates dictated the use of an auger sampling method.

SAMPLING METHOD

The auger sampling method, hereafter referred to as the "Official Purchase Method" is given in the following flowsheet, accompanied by a narrative description.

FLOWSHEET OF "OFFICIAL PURCHASE METHOD"



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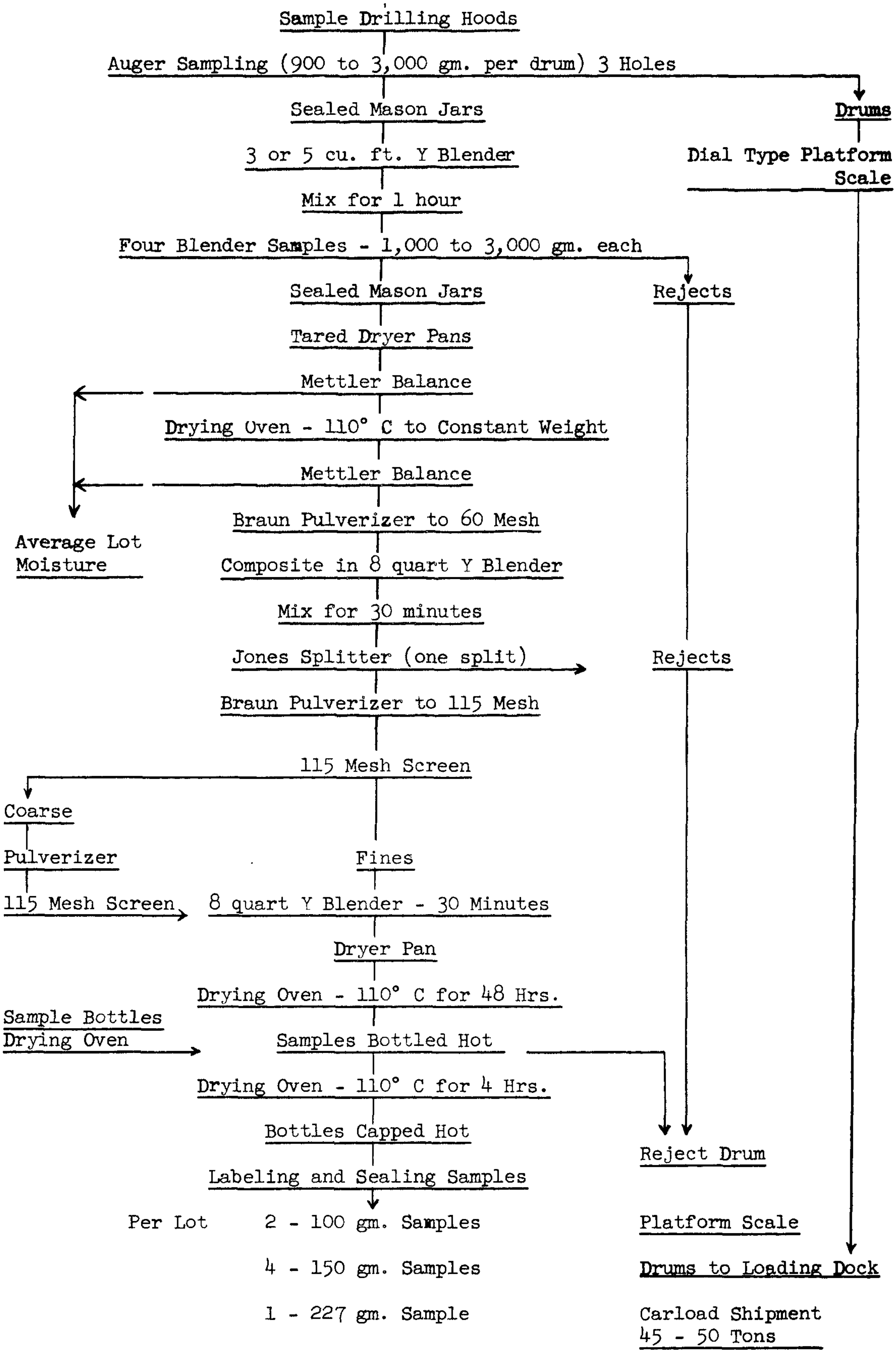
(1) Valent, Dorothy and Gessiness, Bernard, "The Effect of the Moisture Determination on the Assay of Uranium Ore Concentrates". National Lead of Ohio (1957)
Ziegler, W. A., "Evaluation of the Weldon Spring Sampling Plant", Mallinckrodt Chemical Works (1958)

FLOWSHEET (Cont'd)



The description of the "Official Purchase Method" is as follows:

1. Uranium Mill Products are packaged in 55 gallon removable head steel drums with net weight of contents between 300 and 1300 pounds. Shipments are by truck and rail, and are received at the unloading dock, then placed in covered storage until sampled. A lot consists of 10 to 77 drums, having a wet weight of 4000 to 60,000 pounds.
2. The drums are weighed, and gross and tare weights recorded. A 2000 pound dial type platform scale is used for weighing and weights are read to the nearest one-half (1/2) pound.
3. The drums are lined up in front of the sample drilling hoods, the drum lids are removed and auger samples are taken from each drum by means of an electric drill with a 36 inch by 1 3/16 inch ship's auger attached. The three auger samples from each drum are caught in a specially constructed stainless steel pan and immediately placed in a one-half (1/2) gallon Mason jar and sealed. Nine hundred to 3000 grams of sample are so taken from each drum, the quantity varying with the density of the material sampled.
4. When the auger sampling of a lot has been completed, the drum heads are replaced and the drums again weighed and moved to the outdoor loading dock. The individual samples are then quickly emptied into either a three or five cubic foot Patterson-Kelley Y blender fitted with an intensifier bar. The size blender used depends on the total volume of the composite lot sample. The blender is then closed and the sample mixed for one hour.
5. The blender is stopped in a vertical position, a large sample bucket placed under the outlet, and the outlet gate opened. Four blender samples of 1000 to 3000 grams each are taken with a scoop as quickly as possible from the falling stream. The bottom gate of the blender is closed between the taking of each sample. These samples are placed immediately in individual Mason jars and sealed. The remainder of the composite sample is placed in the lot rejects drum.
6. The sealed samples are taken to the moisture weighing room. The samples are placed in tared drying pans, weighed and dried at 110° C. This drying is continued a minimum of 48 hours or until the samples cease to lose weight or begin to gain in weight. The percent weight losses shown by the four samples are averaged and used as the lot moisture.
7. After drying, the four samples are taken to the bucking room, ground through a Braun pulverizer to about 60 mesh, combined and mixed for thirty (30) minutes in an eight (8) quart Patterson-Kelley Y blender. The sample is split once in a Jones splitter and screened through a 115 mesh screen, with any oversize material being reground and screened. The combined sample is mixed for thirty (30) minutes in an 8 quart Patterson-Kelley Y blender. The blended sample is placed in a dryer pan and dried for 48 hours at 110° C. Clean sample bottles are put in a dryer and dried for 30 minutes at 110° C. The sample is removed from the oven and 2 - 100 gram, 4 - 150 gram and 1 - 227 gram samples are weighed into the sample bottles as rapidly as possible. The uncapped bottles are immediately replaced in the drying oven for four (4) hours

at 110° C., then capped hot, sealed and labeled. The samples are distributed as follows: Two to LPI lab, one to vendor and one to Feed Material Plant. The rest are held in reserve for possible umpire or in case of breakage.

8. Rejects from pulping are returned to the same lot rejects drum as above. The total rejects from the lot are then put in a tared, empty thirty (30) gallon drum, weighed, and placed in outdoor storage with the lot from which they came.

9. Shipments are made by rail from the outdoor loading dock. Carload shipments are from forty (40) to fifty (50) tons.

10. All sampling equipment is kept as clean as possible. All glass jars and pans are thoroughly washed after use. Grinding room equipment is blown out with compressed air and washed. Work benches and hoods are frequently washed.

    Dust collected is saved and placed in storage drums. The amount is small. Paper used is burned in an incinerator and the ashes put in a storage drum.

11. Safety measures for men working on uranium mill products include clean caps, coveralls, and cotton gloves daily. Leather gloves are also provided. Regular wash-up and bathing periods are required daily. Respirators are required at all locations where dusting may occur. Dust hoods are provided where possible. Medical examinations are made on these workmen every twelve months and frequent urine analyses for uranium are made.

EVALUATION OUTLINE AND REQUIREMENTS

There have been investigations of certain steps of the auger sampling method, but as far as we know, no thorough step-wise evaluation of the total auger method of sampling uranium concentrates has been carried out.

The sampling method as used at Grand Junction was thoroughly evaluated in a step-wise manner by evaluating each of the following operations:

1. Weighing.
2. Taking of the primary sample.
3. Moisture determination.
4. Sample preparation.
5. Analysis for U_3O_8.

All tests were carried out using the regularly received concentrates and were performed by the operating personnel using standard plant equipment and techniques for both the sampling plant and laboratory.

Because there were no uranium concentrate standards which could be used, another statistically valid method for determining the accuracy of the sampling method and the analytical method had to be used. The test of the accuracy was performed by use of two different and independent methods of measurement of the sampling and analytical steps under investigation. Whenever the two methods show no significant difference the operation under study was considered accurate.

Precision measurements were obtained by pooling the variances of a series of duplicate or replicate measurements.

The data so obtained was analyzed by acceptable statistical methods such as the "t" test, standard deviation, etc. All decisions were made at the 95% confidence level. Whenever the significance level of a set of tests was between 90% and 99%, we continued by sequential testing where possible until a firm decision could be made. The results of the evaluation are as follows.

WEIGHING

The initial weighing of the lot was evaluated as follows:

The uranium concentrates are received in 55 gallon steel drums and a typical lot contains 44 drums and weighs 17,000 to 45,000 lbs. gross. The scale used to obtain the gross weight of the lot is a Howe dial scale of 2000 pound capacity with graduations of 0.5 pound. Each drum is individually weighed and recorded to the nearest 0.5 pound. The accuracy of the scale was not determined, rather the accuracy was and is maintained by the use of NBS Class C tolerance check weights. The scale was checked before and after the weighing of a lot, using the check weights, whose total weight was in the range of the drums of the lot. The results of triplicate weighings of four lots of concentrates is shown in Table I.

Table I

Precision of Weighing

Lot No.		No of Drums	Lot Weight in Pounds First Weighing	 Second Weighing	 Third Weighing
Uranium Reduction	198	30	17,557.0	17,565.0	17,557.0
Uravan	598	27	17,254.0	17,256.5	17,258.0
Anaconda	A1036	33	11,875.5	11,871.5	11,868.0
Anaconda	A1037	33	12,815.5	12,819.5	12,813.0

Standard Deviation = 3.5 pounds per lot.

The standard deviation is 3.5 pounds and converting to a 10,000 pound lot basis, the standard deviation is 2.4 pounds. Later work performed on 17 lots, whose average number of drums was 40% greater than the lots in Table I, yielded a standard deviation of 1.7 pounds on the basis of a 10,000 pound lot. Because the lot sizes vary considerably the results are weighted to a 10,000 pound lot basis for purposes of comparison. The tares of the drums were obtained by taring, before filling, on a 75 pound capacity Howe dial scale which has a standard deviation of 0.08 pounds per drum. This error in taring was too small to be of any consequence in the weighing of a lot.

Before continuing with the evaluation of the "taking of the primary sample", the reasons for drilling three holes in a drum at predetermined poisitions should be clarified.

Three holes are drilled per drum rather than one or two because of the variations in % H_2O and % U_3O_8 within a drum. The possible advantage gained by drilling more than three holes is not equal to the increased man-hours required for the additional holes. A measure of the variation in values between the three holes was obtained by drilling 5 drums of each of 5 typical types of uranium concentrates. Each hole sample was dried for % H_2O and analyzed for U_3O_8. The statistical results are as follows: The units are in % U_3O_8, wet basis.

		Standard Deviations	
Lot No.		Single Hole	Ave. of 3 Holes
Dawn Mining	6	0.135	0.078
Anaconda	A1166	0.672	0.388
Uranium Reduction	316	0.485	0.280
Uravan	650	0.509	0.294
Rare Metals	74	0.290	0.167

The placement of the three holes, A, B, and C are predetermined as shown in the following Diagram No. 1.

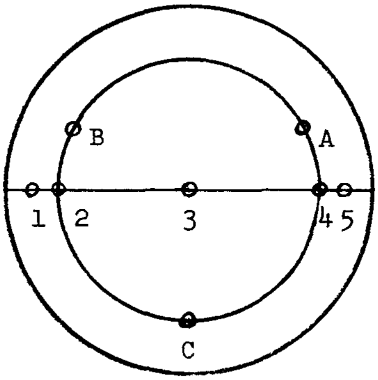


Diagram No. 1

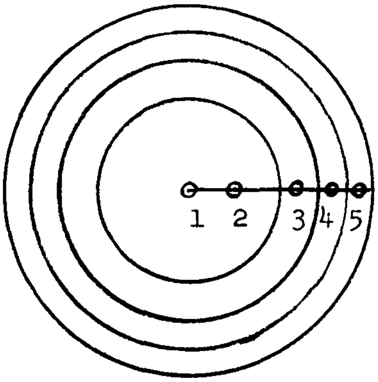


Diagram No. 2

The inner circle and annular rings of each circle are equal in area.

As can be seen by Diagram No. 1, the area and subsequently the volume of the drum is divided into two equal portions by the concentric circle. The holes, A, B, and C are drilled on this circle in order to sample the drum by volume half way from the center to the outside. The reason for this is that the filling of the drums at the uranium mills often results in lateral segregation of particle size. This lateral particle size segregation can also result in lateral segregation of values to varying degrees.

The existence of lateral segregation of values in uranium concentrates was determined as follows. Five drums of four typical lots of concentrates were drilled in the positions 1, 2, 3, and 5 as shown in Diagram No. 2. Each hole sample was dried for % H_2O and analyzed for U_3O_8. Then an additional eight drums of Uranium Reduction Lot No. 579 were drilled in the positions 1, 2, 3, 4 and 5 as shown in Diagram No. 1. The results were as follows:

Lot No.		Average % U_3O_8, Wet Basis Position Numbers 1	2	3	5
Dawn Mining	20	74.77	74.59	75.50	75.70
Anaconda	A1298	78.23	78.18	78.34	78.54
Uranium Reduction	399	84.87	84.97	85.14	84.84
Vitro	82	86.85	86.78	86.89	86.72

Lot No.		Position Numbers 1	2	3	4	5
Uranium Reduction	579	84.40	84.39	83.80	84.52	84.41

F Test for Significant Difference
Between Lateral Positions

Lot No.		F Number	Level of Significance
Dawn Mining	20	3.38	94 %
Anaconda	A1298	2.35	86 %
Uranium Reduction	399	2.21	84 %
Vitro	82	0.34	20 %
Uranium Reduction	579	5.28	99 %

Further proof that the sampling of the drums in these predetermined positions is obtaining a representative sample is shown in the evaluation of the "taking of the primary sample".

TAKING OF PRIMARY SAMPLE

The next phase of the auger sampling method which we evaluated was the accuracy of "taking of the primary sample." We determined the ability of the auger method to obtain a representative sample by sampling the same drums by the "Auger Method" and by a different and independent method of sampling.

The "Auger Method" is the "Official Purchase Method" with a single drum being carried through the procedure.

The independent method of sampling was what we called the "Ten Cubic Foot Blender Method". The largest "Y" blender available at Grand Junction has a capacity of ten cubic feet and will easily accommodate the contents of a 55 gallon drum. It is equipped with an intensifier bar.

Sampling by the "Ten Cubic Foot Blender Method" was performed as follows: Following the auger sampling of a drum chosen at random the contents of the drum were transferred to the blender and blended one hour. As the blended material flowed from the blender, four samples were taken by uniformly cutting the stream. These four samples were transferred to Mason jars and sealed. The size of each blender sample was taken so as to be nearly equal to that obtained by the auger.

The samples from the ten cubic foot blender were dried for the moisture determination and analytical samples prepared as given in the "Official Purchase Method". The auger samples of a drum were processed side by side with the blender samples from that drum so as to exclude errors not pertinent to the test.

The statistical results of these tests are given in Table II.

Table II

Accuracy of Taking the Primary Sample by the Auger

Vendor	No. Drums Tested	Significance Level	Lbs. U_3O_8 Contained Diff. Auger Less Blender	
			Average Per Drum	Ave. 44 Drum Lot*
Anaconda	12	58.5	+ 0.07	+ 0.46
Kerr McGee	12	15.1	- 0.08	- 0.53
Uranium Reduction	16	66.8	- 0.93	- 6.17
Uravan	12	50.3	+ 0.05	+ 0.33
Vitro	16	60.8	- 0.21	- 1.39

*Extrapolation

$$\text{Difference per drum times } \sqrt{\text{No. Drums in lot}} = \text{Difference per lot.}$$

The extrapolation was performed to obtain a measure of the differences that could be expected for a full lot.

The two methods of taking the primary sample do not differ significantly. The sample from the blender mix is a representative sample and therefore, the auger sampling method is also obtaining a representative sample from the drum. The efficiency of the blending is well illustrated by the low moisture content differences found between the blender samples. (See Table IV)

The precision of the auger sampling operation was determined by performing duplicate samplings of a series of drums chosen at random. The duplicate samples were obtained by augering and dried. Analytical samples were prepared as given in the "Official Purchase Method".

The duplicate samples were carried through the drying and sample preparation side by side in order to minimize the errors not pertinent to the test.

The results are given in Table III.

MOISTURE DETERMINATION

The moisture determination is the next sampling step which was evaluated.

The precision of the moisture determination was obtained by pooling the variances of sets of four identical samples. The samples are the samples taken from the discharge stream of the blender following the blending of the auger samples as described in the "Official Purchase Method". The data are actual purchase results. The results are given by moisture levels in Table IV.

Table III

Precision of Auger Sampling

Vendor	Number of Drums Tested	Standard Deviation in Pounds U_3O_8 Contained Single Drum	 44 Drum Lot*	 Per 10,000 Lb. Lot*
Anaconda	12	0.20	1.33	1.03
Kerr McGee	12	0.52	3.45	2.07
Uranium Reduction	16	1.05	6.96	4.14
Uravan	12	0.31	2.06	1.50
Vitro	16	0.51	3.38	1.89

*Extrapolation

$$\text{S of n drum lot} = \text{S of single drum} \times \sqrt{\text{No. Drums in lot}}$$

$$\text{S for a 10,000 lb. lot} = \text{S of single drum} \times \sqrt{\text{No. Drums in a 10,000 lb. lot}}$$

The standard deviation for a single drum was extrapolated to a typical 44 drum lot basis and found to be not over 7.00 pounds of U_3O_8 contained for a typical lot. The standard deviation for a single drum was extrapolated to a 10,000 pound lot and found to be not over 4.2 pounds U_3O_8 contained.

Table IV

Precision of Moisture Determination

% H_2O Level	Average % H_2O	No. of Lots	Degrees of Freedom	Standard Deviation % H_2O Absolute
0.0-1.0	0.336	10	30	0.012
1.0-2.0	1.468	10	30	0.022
2.0-3.0	2.317	9	27	0.028
3.0-4.0	3.315	9	27	0.023
4.0-5.0	4.689	8	24	0.029
5.0-6.0	5.458	7	21	0.051
6.0-7.0	6.457	6	18	0.027
7.0-8.0	7.579	4	12	0.039
8.0-9.0	8.294	1	3	0.029
9.0-10.0	9.634	1	3	0.017

The precision of this measurement is good and no significant change occurs in the % H_2O determination precision as the H_2O content increases.

The accuracy of the moisture determination was much more difficult to determine. We used the principle of two independent separate methods for our evaluation of this phase of the sampling method.

The two methods used were the "Official Purchase Method" and the "Pipe" method.

As can be seen by the flowsheet, the accuracy of the moisture determination depends on the ability of the sampling method to produce a representative sample for analysis which is in the same state of dryness as existed in the moisture samples at the end of the moisture determination. When this is accomplished the laboratory is reporting a % U_3O_8 which is the same as existed in the dried moisture samples and then by using the % H_2O and % U_3O_8 with the net wet weight of the lot we have the true pounds U_3O_8 contained for the lot.

The redrying of the sieved and blended sample as given in the "Official Purchase Method" is designed to return the final sample to the same state of dryness as existed at the end of the moisture determination. For this reason we included all sample preparation steps, which followed the moisture determination, in the determination of the accuracy of the moisture determination.

The "Pipe" method for determining this accuracy is as follows: Immediately following the taking of the four moisture samples from the blended drum samples (Official Purchase Method) the rejects were leveled and five 1 1/2" x 9" pipes of thin wall electrical conduit were inserted one at each corner and one in the center of the reject pan. The pipes were immediately stoppered, and by means of an angle spatula, the contents of each pipe was transferred to a tared Erlenmeyer flask and stoppered. The flask and contents were weighed and the total content of the flask, between 100 and 200 grams of uranium concentrates, was dissolved in mixed acids.

Liquid samples were weighed from the pipe sample solutions and analyzed for U_3O_8.

This test was performed on 23 lots of uranium concentrates representing 14 different vendors and all types of uranium concentrates received at Grand Junction.

The results of the statistical analysis are given in Table V.

Table V

Accuracy of the Moisture Determination
Found by Comparing Two Independent Methods
"Official Purchase and Pipe"

	Pounds U_3O_8 Contained Average Difference Official Less Pipe	T Number	Level of Significance
Unweighted	- 3.57	1.037	68.8 %
Weighted to 10,000 lb. Net wet lot	- 0.46	0.161	12.6 %

The average net wet weight for the 23 lots was 13,640 pounds.

There is no significant difference between the "Official Purchase Method" and the "Pipe" method of determining the moisture. These results show that we are obtaining an analytical sample which is at the same degree of dryness that existed at the end of the moisture determination.

SAMPLE PREPARATION

The sample preparation step of the "Official Purchase Method" has been included in the accuracy of the moisture determination. Nevertheless, the effects on the analytical sample of possible heating during pulverizing and of redrying of the ground and blended sample was investigated.

The test procedure was as follows. A sample of the blender rejects was dried in the usual manner. Immediately following the final weighing of the moisture determination 100 to 200 grams of the dried sample was quickly transferred by pipe sampling to a tared Erlenmeyer flask and stoppered.

The flask and contents were weighed and the total contents of the flask were dissolved in mixed acids. Liquid samples were weighed from the pipe sample solutions and analyzed for U_3O_8. An analytical sample was prepared from the dried sample remaining in the drying pan, as given in the "Official Purchase Method". The % U_3O_8 was determined on this prepared sample.

The results of the test are given in Table VI.

Table VI

Accuracy of the Sample Preparation

Percent U_3O_8 in the Prepared Analytical Sample Versus
The % U_3O_8 in the Sample Before Preparation

Number of lots tested	19
Difference (after preparation less before preparation	+ 0.037% U_3O_8
Significance Level	82.5%

As was found in the "Pipe" tests, the sample preparation and redrying of the ground and screened sample have no slanting effect on the analytical sample.

Probably by this time you have a question in your mind concerning the validity of blending the drum samples as received for both the moisture and analytical samples. Theoretically the samples should be composited on a weighted basis. We have found by inspection of the weights of the drum samples (auger sample) from many lots that the ship's auger as used at Grand Junction obtains a nearly weighted sample (See Appendix Table XIA) The actual auger sample for 25 lots of various types of concentrates differed from the weighted sample by an average of 5.9 % of the sample.

Blending of the drum samples as received effected considerable savings in cost. The blending of the analytical sample without compositing reduced the labor cost of this portion of the sample preparation by 90%. The blending of the drum samples for the moisture sample rather than drying each individual drum sample saved 80% of the labor cost of the moisture determination.

ANALYTICAL METHOD

The fifth step evaluated was the U_3O_8 determination. The analytical method used for the determination of U_3O_8 in uranium concentrates is the "Zinc Reductor" method. This method of analysis consists of a mixed acid digestion, sulfide and cupferron separations in sulfuric acid solution, reduction in a zinc reductor and titration with potassium dichromate solution. Sodium diphenylamine sulfonate is used as the titration indicator. This method is an adaptation of the method in use at the National Bureau of Standards.

The accuracy of the analytical method was determined by the principle of different and independent methods. The second method consists of a mixed acid digestion, phosphate precipitation, cupferron separation, reduction in a lead reductor and titration with ceric sulfate using ferroin as the indicator. This method of analysis was obtained from the Mallinckrodt Chemical Works at Weldon Spring, Missouri. In the latter method the uranium is completely precipitated by the phosphate in the first separation, while in the "Zinc Reductor" method the uranium remains in solution throughout the procedure.

Four samples from each of nine lots of representative uranium concentrates were weighed out and two samples of each set analyzed by each method. The duplicates were averaged and the averages compared by the "t" test. The results are given in Table VII.

Table VII

Accuracy of the U_3O_8 Determination

Lot No.		% U_3O_8 Difference LPI Procedure Less Independent Procedure
Texas Zinc	97	- 0.01
Durango	585	+ 0.02
Uranium Reduction	572	- 0.04
Uranium Reduction	573	- 0.08
Vitro	149	+ 0.15
Western Nuclear	76	- 0.08
Mines Development	107	- 0.09
Dawn Mining	56	0.00
Phillips	24	- 0.15

Average difference = - 0.03 Significance level = 68.7 %

The "t" test indicates no slant between the methods and the significance level was found to be 68.7%. Therefore, the "Zinc Reductor" method is accurate. It should be mentioned here that the routine standardization of the titrating solution is carried out using standard uranium black oxide, U_3O_8. This standardization operation encompasses the aliquoting, reduction and titrating operation of our analytical method, so, the accuracy test primarily covered the weighing, acid digestion and the two separation operations of the procedure.

The precision of the analytical method is under constant surveillance and can be reported in two ways. The first is the precision found by pooling the variances of duplicate determinations which are performed on the same day by different analysts. Second is the precision found by analyzing identical samples over several months time by several analysts. The latter measure of precision includes analyst drift and the other long term changes. The short term precision, utilizing the routine purchase assays which are performed in duplicate on the same day by different analysts, has a standard deviation for the analytical method of 0.C3 % U_3O_8 absolute. The long term precision measurements which covered 6 months and involved 12 sets of 21 identical samples of 11 representative uranium concentrates showed a standard deviation of 0.09 % U_3O_8 absolute. Table VIII.

Table VIII

Precision of the U_3O_8 Determination

	Number of Determinations	Standard Deviation % U_3O_8 Absolute
Short Interval Precision	>300	0.027
Long Interval Precision	486	0.085

Our laboratory reports for purchase purposes the average of duplicate U_3O_8 determinations. The standard deviation for an average of 2 determinations is 0.060 % U_3O_8 absolute for the long interval.

An indication of the accuracy and precision available by our U_3O_8 analytical method is the following comparisons with other analytical laboratories which reported the % U_3O_8 found in replicate samples sent to them. Table IX.

Table IX

Comparisons Between the Grand Junction Laboratory and Other Laboratories As Reported in the Fiscal Year 1958

Comparison Basis - Purchase Assays % U_3O_8 Dried at 110° C.

Comparison Laboratory	Number of Comparisons	Average Difference G.J. Less Comparison	Minimum Detectable Diff.
A	408	+ 0.058	0.01
B	289	- 0.059	0.02
C	128	- 0.140	0.03
D	71	- 0.159	0.05
E	34	- 0.139	0.06
F	160	- 0.043	0.02

A, B, C, D and E are vendor laboratories. Laboratory No. F is the National Bureau of Standards. All umpire work on uranium concentrates received at Grand Junction is performed at NBS, and Lucius Pitkin, Inc. won 93.8% of the umpires in fiscal year 1958.

As can be seen, the agreement between laboratories is exceptionally good. The analyses reported by the Grand Junction laboratory and the N.B.S. laboratory were performed on the average of four months apart, and as is well known, in a widely different climate. The climatic difference is also aggravated by seasonal differences due to the elapsed times between analysis.

After evaluating the various steps of the sampling and analytical operation the precision of the overall operation was obtained. This included all steps from the weighing of the lot thru the analysis of the dried sample. This overall precision is not a day to day precision measurement, but the precision obtained from performing the duplicate measurements of each step, side by side, on the same days. Eighteen lots were sampled in duplicate and the standard deviation of the sampling and analysis combined was found to be 4.94 lbs. U_3O_8 contained on the basis of a 10,000 lb. net wet lot weight (See Appendix Table VIIA). The standard deviation of the overall operation of these lots is equivalent to 0.07% of the pounds of U_3O_8 contained in the lots.

The costs, as well as the accuracy and precision, must be considered in the evaluation of a sampling and analytical operation.

SAMPLING COSTS

The sampling plant employs 2 supervisors, 4 clerical personnel and 24 plant operators working only the day shift on a five day week. Allowing for change time, clean-up time, etc., the sampling plant sampled, on the average, of one drum of concentrates every 1.36 minutes for the month of March, 1959. The sampling plant has a capacity of 50% greater than the present volume of concentrates being received.

The costs given here are total costs which include all direct and indirect operating and administrative costs.

For March, 1959, the costs per 100 pounds of net wet concentrate sampled were $0.57, $0.45 and $0.05 for the sampling, analysis and the shipping, respectively. The sampling also includes the lot moisture determination.

Estimated Future costs for the sampling, analysis and shipping of uranium concentrates have been calculated. These costs are $0.51, $0.39 and $0.05 for the sampling, analysis, and shipping, respectively.

The reduction in these costs is due to an estimated 5% increase in concentrate receipts and a reduction in administrative costs because of additional contractor activities.

ACKNOWLEDGEMENT

The author wishes to thank the personnel of Lucius Pitkin, Inc. and the Grand Junction Operations Office of the U. S. Atomic Energy Commission for their comments and helpful suggestions.

The Grand Junction Sampling Plant and Laboratory are to be commended for their cooperation in carrying out the physical operations of the Evaluation Program.

APPENDIX

Table IA

Precision of Weighing
Source of Data is the MCW Check Lots
Which Were Sampled at Grand Junction in Late
January, 1959 and February, 1959

Lot No.		No. of Drums	Net Wet Weights - Pounds		
			1	2	3
Texas Zinc	99	44	29,127.5	29,130.5	29,133.0
" "	100	44	29,388.0	29,386.5	29,393.5
" "	101	44	28,488.0	28,481.5	28,490.0
" "	102	22	15,804.0	15,807.5	15,804.5
" "	103	44	31,093.5	31,098.5	31,107.5
Mines Dev.	106	77	44,806.6	44,818.1	44,822.6
" "	107	39	20,951.0	20,951.0	20,951.0
" "	108	77	40,772.9	40,769.4	40,785.4
" "	109	34	17,927.2	17,923.2	17,935.2
Phillips Petroleum	22	44	44,015.4	44,022.4	44,023.4
" "	23	44	44,044.5	44,038.5	44,043.5
" "	24	44	44,042.7	44,042.2	44,057.2
Rifle	R-64	55	39,627.2	39,611.7	39,623.7
"	R-65	33	22,884.5	22,882.5	22,886.5
Durango	586	22	26,341.5	26,342.5	26,344.0
"	587	20	24,028.5	24,028.5	24,029.5
Lucky Mc	73	48	30,038.4	30,022.4	30,034.4
Ave. Per Lot		43	31,375.4	31,373.9	31,380.3

S = 5.5 pounds for 31,375 pound lot.

S = 1.70 pounds on basis of 10,000 pound net wet lot.

The S for a lot on a 10,000 pound basis was obtained as follows:

The No. 1 weighing was arbitrarily set at 10,000 lbs. and the weights of Nos. 2 & 3 on a 10,000 pound lot basis were obtained by the following equations.

$$\text{No. 2} = \frac{\text{Wt. No. 2}}{\text{Wt. No. 1}} \times 10{,}000 \quad \text{and} \quad \text{No. 3} = \frac{\text{Wt No. 3}}{\text{Wt. No. 1}} \times 10{,}000.$$

The variances of the sets of three weighings were pooled to obtain the S of 1.70 pounds per 10,000 pound lot.

Table IIA

Accuracy of Taking Primary Sample by the Auger

POUNDS U_3O_8 CONTAINED

Anaconda A1510 Drum No.	Duplicate Auger Samples A-1	A-2	Average A-1 & A-2	Blender	Ave. A-1 & A-2 Less Blender
2	291.93	292.43	292.18	292.33	- .15
10	289.13	288.97	289.05	289.17	- .12
15	269.44	269.17	269.30	268.75	+ .55
28	274.06	274.22	274.14	274.02	+ .12
			1124.67	1124.27	+ .40
Anaconda A1521					
10	280.36	280.57	280.46	280.61	- .15
18	271.67	272.10	271.88	271.80	+ .08
25	270.25	269.83	270.04	269.65	+ .39
29	294.28	294.05	294.16	294.04	+ .12
			1116.54	1116.10	+ .44
Anaconda A1523					
12	306.64	306.80	306.72	306.41	+ .31
19	318.14	317.92	318.03	318.51	- .48
47	332.06	332.11	332.08	331.78	+ .30
63	279.88	279.62	279.75	279.87	- .12
			1236.58	1236.57	+ .01
	Total All Drums				+ .85
	Ave. All Drums				+ .0708

Table IIA (Cont'd)

Accuracy of Taking Primary Sample by the Auger

POUNDS U_3O_8 CONTAINED

Kerr McGee 166

Drum No.	Duplicate Auger Samples		Average A-1 & A-2	Blender	Ave. A-1 & A-2 Less Blender
	A-1	A-2			
8	328.52	328.47	328.50	328.07	+ .43
10	332.23	331.59	331.91	330.97	+ .94
12	349.97	349.48	349.72	349.41	+ .31
37	421.69	421.53	421.61	423.42	- 1.81
			1431.74	1431.87	- .13
Kerr McGee 167					
7	546.20	546.38	546.29	548.14	- 1.85
9	564.00	563.72	563.86	564.90	- 1.04
22	357.83	358.02	357.92	357.14	+ .78
48	660.62	660.45	660.54	657.55	+ 2.99
			2128.61	2127.73	+ .88
Kerr McGee 168					
7	759.99	759.21	759.60	759.56	+ .04
16	732.06	732.19	732.12	731.60	+ .52
19	726.05	727.57	726.81	728.99	- 2.18
25	558.98	557.33	558.16	558.28	- .12
			2776.69	2778.43	- 1.74
	Total All Drums				- .99
	Ave. All Drums				- .0825

Table IIA (Cont'd)

Accuracy of Taking Primary Sample by the Auger

POUNDS U_3O_8 CONTAINED

U.R.C 540

	Duplicate Auger Samples		Average		Ave. A-1 & A-2
Drum No.	A-1	A-2	A-1 & A-2	Blender	Less Blender
1	605.41	607.10	606.26	607.24	- .98
20	562.31	561.51	561.91	560.42	+ 1.49
26	617.57	616.73	617.15	615.47	+ 1.68
45	612.73	613.93	613.33	615.28	- 1.95
			2398.65	2398.41	+ .24
U.R.C. 541					
4	578.82	579.42	579.12	581.65	- 2.53
6	522.70	522.56	522.63	522.79	- .16
30	507.75	510.07	508.91	514.39	- 5.48
54	574.72	575.58	575.15	577.95	- 2.80
			2185.81	2196.78	-10.97
U.R.C. 546					
23	511.46	510.22	510.84	510.62	+ .22
30	473.03	471.73	472.38	477.62	- 5.24
36	480.74	482.01	481.38	488.79	- 7.41
50	374.72	372.91	373.82	375.81	- 1.99
			1838.42	1852.84	-14.42
U.R.C. 571					
10	523.27	522.96	523.12	521.78	+ 1.34
18	572.29	574.64	573.46	574.29	- .83
25	531.77	533.52	532.64	531.30	+ 1.34
29	588.06	585.60	586.83	578.36	+ 8.47
			2216.05	2205.73	+10.32
	Total All Drums				-14.83
	Ave. All Drums				- .9269

APPENDIX (Continued)

Table IIA (Cont'd)

Accuracy of Taking Primary Sample by the Auger

POUNDS U_3O_8 CONTAINED

Uravan 759

Drum No.	Duplicate Auger Samples A-1	Duplicate Auger Samples A-2	Average A-1 & A-2	Blender	Ave. A-1 & A-2 Less Blender
10	347.14	347.59	347.36	347.27	+ .09
18	303.93	304.50	304.22	304.22	00
25	344.75	343.80	344.28	344.28	00
29	364.46	364.42	364.44	364.27	+ .17
			1360.30	1360.04	+ .26
Uravan 761					
12	279.16	278.98	279.07	278.80	+ .27
19	285.78	285.78	285.78	285.78	00
47	319.84	319.43	319.64	319.51	+ .13
63	366.17	365.79	365.98	365.55	+ .43
			1250.47	1249.64	+ .83
Uravan 762					
2	365.88	365.74	365.81	366.15	- .34
10	326.68	326.13	326.40	326.17	+ .23
15	362.71	362.43	362.57	362.66	- .09
28	312.94	313.26	313.10	313.43	- .33
			1367.88	1368.41	- .53
	Total All Drums				+ .56
	Ave. All Drums				+ .0467

Table IIA (Cont'd)

Accuracy of Taking Primary Sample by the Auger

POUNDS U_3O_8 CONTAINED

Vitro 136	Duplicate Auger Samples		Average		Ave. A-1 & A-2
Drum No.	A-1	A-2	A-1 & A-2	Blender	Less Blender
14	575.41	575.21	575.31	575.08	+ .23
44	731.86	733.75	732.80	733.53	- .73
47	714.31	714.16	714.24	714.27	- .03
58	641.74	641.40	641.57	642.37	- .80
			2663.92	2665.25	- 1.33
Vitro 138					
12	743.16	743.42	743.29	743.51	- .22
19	602.32	602.65	602.48	602.62	- .14
47	787.66	786.88	787.27	788.01	- .74
63	652.96	652.96	652.96	653.30	- .34
			2786.00	2787.44	- 1.44
Vitro 139					
1	572.08	572.27	572.18	572.16	+ .02
20	666.47	666.05	666.26	666.30	- .04
26	730.63	730.63	730.63	731.22	- .59
45	613.84	613.07	613.46	613.92	- .46
			2582.53	2583.60	- 1.07
Vitro 149					
11	535.76	536.79	536.28	537.06	- .78
26	530.36	530.70	530.53	527.64	+ 2.89
32	506.16	504.78	505.47	506.47	- 1.00
33	487.04	487.16	487.10	487.65	- .55
			2059.38	2058.82	+ .56
	Total All Drums				- 3.28
	Ave. All Drums				- .2050

Table IIIA

Precision of Auger Sampling of Single Drums
(See A-1 and A-2 of Table IIA)

Lot No.	Drum No.	Lbs. U_3O_8 Contained A-1 less A-2
Anaconda		
A1510	2	- 0.50
"	10	+ 0.16
"	15	+ 0.27
"	28	- 0.16
A1521	10	- 0.21
"	18	- 0.43
"	25	+ 0.42
"	29	+ 0.23
A1523	12	- 0.16
"	19	+ 0.22
"	47	- 0.05
"	63	+ 0.26
	S = 0.20	Lbs. U_3O_8 Contained
Kerr McGee		
166	8	+ 0.05
"	10	+ 0.64
"	12	+ 0.49
"	37	+ 0.16
167	7	- 0.18
"	9	+ 0.28
"	22	- 0.19
"	48	+ 0.17
168	7	+ 0.78
"	16	- 0.13
"	19	- 1.52
"	25	+ 1.65
	S = 0.52	Lbs. U_3O_8 Contained

Lot No.	Drum No.	Lbs. U_3O_8 Contained A-1 less A-2
Uravan		
759	10	- 0.45
"	18	- 0.57
"	25	+ 0.95
"	29	+ 0.04
761	12	+ 0.18
"	19	0.00
"	47	+ 0.41
"	63	+ 0.38
762	2	+ 0.14
"	10	+ 0.55
"	15	+ 0.28
"	28	- 0.32
	S = 0.31	Lbs. U_3O_8 Contained

APPENDIX (Continued)

Table IIIA (Cont'd)

Precision of Auger Sampling of Single Drums
(See A-1 and A-2 of Table IIA)

Lot No.	Drum No.	Lbs. U_3O_8 Contained A-1 less A-2	Lot No.	Drum No.	Lbs. U_3O_8 Contained A-1 less A-2
Vitro			Uranium Reduction		
136	14	+ 0.20	540	1	- 1.69
"	44	- 1.89	"	20	+ 0.80
"	47	+ 0.15	"	26	+ 0.84
"	58	+ 0.34	"	45	- 1.20
138	12	- 0.24	"	4	- 0.60
"	19	- 0.33	"	6	+ 0.14
"	47	+ 0.78	"	30	- 2.32
"	63	0.00	"	54	- 0.86
139	1	- 0.19	546	23	+ 1.24
"	20	+ 0.42	"	30	+ 1.30
"	26	0.00	"	36	- 1.27
"	45	+ 0.77	"	50	+ 1.81
149	11	- 1.03	571	10	+ 0.31
"	26	- 0.34	"	18	- 2.35
"	32	+ 1.38	"	25	- 1.75
"	33	- 0.12	"	29	+ 2.46
	S = 0.51	Lbs. U_3O_8 Contained		S = 1.05	Lbs. U_3O_8 Contained

APPENDIX (Continued)

Table IVA

Accuracy of the Moisture Determination

		Pounds U_3O_8 Contained		Official Purchase Less Pipe	
Lot No.		Official Purchase	Pipe	Unweighted	Weighted To 10,000 Lbs. Net Wet Lot
Anaconda	A891	7,954.06	7,965.40	- 11.34	- 11.50
"	A932	8,193.22	8,201.25	- 8.03	- 8.00
"	A940	8,483.95	8,485.51	- 1.56	- 1.50
"	A1009	7,603.75	7,593.86	+ 9.89	+ 9.73
"	A1033	7,879.49	7,849.01	+ 30.48	+ 31.50
"	A1049	9,784.06	9,774.86	+ 9.20	+ 7.50
"	A1064	9,946.82	9,950.62	- 3.80	- 3.00
"	A1077	9,285.36	9,282.46	+ 2.90	+ 2.50
"	A1083	8,515.38	8,524.74	- 9.36	- 8.51
Climax	255	15,355.28	15,364.40	- 9.12	- 4.90
Durango	526	12,499.48	12,515.40	- 15.92	- 11.04
Kerr McGee	126	13,051.11	13,070.69	- 19.58	- 11.50
Mines Development	64	9,375.65	9,365.57	+ 10.08	+ 7.51
Naturita	376	12,375.16	12,360.26	+ 14.90	+ 9.95
N.L. Monticello	C-58	12,175.84	12,188.25	- 12.41	- 6.82
" "	A-142	4,582.53	4,557.88	+ 24.65	+ 34.65
Rare Metals	75	7,185.72	7,188.82	- 3.10	- 2.96
Rifle	R-52	13,094.06	13,126.96	- 32.90	- 19.73
Texas Zinc	4	7,959.40	7,950.29	+ 9.11	+ 8.81
UCN Uravan	576	11,860.98	11,860.22	+ 0.76	+ .50
Uranium Reduction	166	12,801.51	12,807.56	- 6.05	- 3.50
Vitro	70	18,878.44	18,907.90	- 29.46	- 11.66
Western Nuclear	24	12,824.47	12,855.96	- 31.49	- 18.51
			Σd_i =	- 82.15	- 10.48
			$\bar{d}$ =	- 3.57	- .46

	t	Sig. Level
Unweighted	1.037	68.8%
Weighted to 10,000 lb. Net wet lot	.161	12.6%

Note: The differences noted for any one vendor is not representative of that product, unless at least 8 or 10 lots of that product are grouped for study.

APPENDIX (Continued)

Table VA

Accuracy of the Sample Preparation

Lot No.		1. % U_3O_8 at End of Moisture Determination	2. % U_3O_8 in Analytical Sample	Diff. 1 Less 2
Anaconda	A1049	83.50	83.55	- 0.05
"	A1064	82.45	82.53	- 0.08
"	A1077	83.75	83.90	- 0.15
"	A1083	81.39	81.39	0.00
"	A1096	83.24	83.24	0.00
"	A1397	82.82	82.77	+ 0.05
"	A1399	83.42	83.42	0.00
"	A1401	81.46	81.56	- 0.10
Texas Zinc	48	77.32	77.46	- 0.14
" "	50	75.73	76.08	- 0.35
Uranium Reduction	233	82.81	82.72	+ 0.09
" "	257	71.17	71.24	- 0.07
" "	260	76.70	76.63	+ 0.07
" "	476	76.42	76.38	+ 0.04
" "	477	74.53	74.49	+ 0.04
Uravan	723	75.02	75.17	- 0.15
"	731	72.18	72.03	+ 0.15
Western Nuclear	45	75.27	75.24	+ 0.03
" "	46	77.34	77.43	- 0.09
			Average	- 0.0374
			t =	1.430
			Significance Level =	82.5 %

Note: At least 8 or 10 lots of a vendor must be grouped for study before an estimate of significant difference can be made for any one vendor's product.

APPENDIX (Continued)

Table VIA

The Accuracy and Precision of the U_3O_8 Determination

Accuracy

Lot No.		Average Determination LPI Method	MCW Method	Difference LPI Less MCW
Durango	585	90.28	90.26	+ 0.02
Dawn Mining	56	79.14	79.14	0.00
Mines Development	107	82.81	82.90	- 0.09
Phillips	24	77.71	77.86	- 0.15
Texas Zinc	97	81.15	81.16	- 0.01
Uranium Reduction	572	85.85	85.89	- 0.04
Uranium Reduction	573	86.80	86.88	- 0.08
Vitro	149	80.51	80.36	+ 0.15
Western Nuclear	76	75.58	75.66	- 0.08

Average = - 0.031

t = 1.080

Significance Level = 68.7 %

The Accuracy and Precision of the U_3O_8 Determination

Precision by Duplicates
(Short Term)

Lot No.		LPI Method 1	LPI Method 2	Diff. 1 Less 2	MCW Method 1	MCW Method 2	Diff. 1 Less 2
Durango	585	90.28	90.29	- 0.01	90.25	90.28	- 0.03
Dawn Mining	56	79.11	79.16	- 0.05	79.17	79.12	+ 0.05
Mines Development	107	82.83	82.79	+ 0.04	82.88	82.93	- 0.05
Phillips	24	77.70	77.72	- 0.02	77.88	77.84	+ 0.04
Texas Zinc	97	81.13	81.17	- 0.04	81.16	81.15	+ 0.01
Uranium Reduction	572	85.86	85.84	+ 0.02	85.83	85.95	- 0.12
Uranium Reduction	573	86.76	86.83	- 0.07	86.87	86.90	- 0.03
Vitro	149	80.51	80.51	0.00	80.40	80.33	+ 0.07
Western Nuclear	76	75.56	75.60	- 0.04	75.58	75.75	- 0.17

LPI S = 0.027 % U_3O_8 absolute

Table VIA (Cont'd)

The Accuracy and Precision of the U_3O_8 Determination

Precision by Duplicates
(Long Term)

Sample No.	No. of Determinations	Average % U_3O_8	% U_3O_8 Absolute Standard Deviation
A	40	82.00	0.077
B	42	78.69	0.074
C	40	83.96	0.115
D	40	84.67	0.106
E	40	65.82	0.093
F	42	70.04	0.037
G	40	60.86	0.060
H	40	82.29	0.070
I	42	85.05	0.092
J	40	74.76	0.066
K	40	76.22	0.118
L	40	68.44	0.081
Total	486		

Pooled standard deviation = 0.085 % U_3O_8 absolute

Standard deviation for an average
of two determinations = 0.060 % U_3O_8 absolute

Table VIIA

Overall Sampling and Analytical Precision

		Pounds U_3O_8 Contained			
Lot No.		Purchase	Purchase Check	Diff. - Purchase Less Purchase Check	Diff. - Weighted to 10,000 Lb. Net Wet Lot
Durango	586	23,415.85	23,424.07	- 8.22	- 3.12
Durango	587	20,667.44	20,632.94	+ 34.50	+ 14.36
Mines Dev.	106	34,580.70	34,590.52	- 9.82	- 2.19
Mines Dev.	107	16,296.93	16,332.96	- 36.03	- 17.20
Mines Dev.	108	31,752.35	31,752.63	- 0.28	- 0.07
Phillips	22	34,208.77	34,201.07	+ 7.70	+ 1.75
Phillips	23	34,143.30	34,164.93	- 21.63	- 4.91
Phillips	24	34,225.58	34,238.34	- 12.76	- 2.90
Lucky Mc	73	23,256.83	23,232.00	+ 24.83	+ 8.27
Texas Zinc	98	22,052.70	22,054.67	- 1.97	- 0.69
Texas Zinc	99	22,367.57	22,370.69	- 3.12	- 1.07
Texas Zinc	100	23,171.60	23,176.97	- 5.37	- 1.83
Texas Zinc	101	22,532.42	22,507.49	+ 24.93	+ 8.75
Texas Zinc	102	12,355.46	12,366.95	- 11.49	- 7.27
Rifle	R-64	31,077.12	31,051.31	+ 25.81	+ 6.51
Rifle	R-65	18,680.14	18,675.26	+ 4.88	+ 2.13
Texas Zinc	103	24,258.78	24,239.11	+ 19.67	+ 6.33
Mines Dev.	109	14,023.21	14,012.15	+ 11.06	+ 6.17

Standard deviation = 4.94 Lbs. U_3O_8 Contained on the basis of a 10,000 Lb. net wet lot.

Table VIIIA

Particle Size and Uniformity of % U_3O_8 and % H_2O

Table A

Particle Size

Lot No.		% Minus 60 Mesh
Uranium Reduction	557	82.0
Dawn Mining	36	19.5
Uravan	734	22.2
Texas Zinc	54	85.5
Homestake N.M.	14	92.8

Table B

Drum to Drum Variation

Anaconda A1085	79.96 % to 84.36 % U_3O_8
(33 Drums)	5.21 % to 8.97 % H_2O

Table C

Within Drum Differences-Maximum Differences Between Any Two of Three Single Auger Samples

Lot No.		No. of Drums Tested	Maximum Differences % U_3O_8	Maximum Differences % H_2O
Rare Metals	74	5	0.38	0.77
Uranium Reduction	316	5	1.00	0.58
Uravan	650	5	1.65	0.24
Anaconda	A1166	5	1.59	0.85
Dawn Mining	6	5	0.34	0.39

Table IXA

Percent Deviation of Actual Drum Sample From Weighted Drum Sample

Method of Calculation:

$$\% \text{ of Lot as Sample} = 100 \times \frac{\text{Sum of Drum Samples}}{\text{Net Wet Lot Weight}}$$

$$\text{Weighted Drum Sample} = \text{Drum Weight} \times \frac{\% \text{ of Lot as Sample}}{100}$$

$$\% \text{ of Deviation} = 100 \times \frac{\text{Actual Drum Sample} - \text{Weighted Drum Sample}}{\text{Actual Drum Sample}}$$

Lot No.		No. of Drums	Ave. Sample Deviation-%
Anaconda	A945	33	5.14
"	A946	33	3.28
"	A948	33	3.42
"	A949	33	4.14
"	A970	33	3.67
"	A978	33	3.15
"	C979	33	3.05
Climax	234	40	9.93
Durango	494	12	2.46
Kerr McGee	126	50	10.84
" "	126	50	9.57
Monticello	A90	55	3.37
Naturita	353	15	6.30
"	354	15	9.18
Rifle	R-28	22	5.70
"	R-31	29	8.38
Rare Metals	36	23	3.11
Uranium Reduction	127	30	6.42
" "	128	30	9.87
" "	129	30	9.59
" "	130	30	5.62
" "	166	30	5.59
" "	169	30	4.87
" "	170	30	4.74
" "	171	30	6.23
	Overall Average		5.9

NONDESTRUCTIVE DETERMINATION OF UO_2 DISTRIBUTION IN UO_2-Zr DISPERSION TYPE FUEL PLATES

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INTRODUCTION

This paper is intended to summarize the experimental work completed, to date, in developing a nondestructive method for the determination of the distribution of uranium in UO_2–Zircaloy-2 dispersion type fuel specimen. The work reported here is limited to an evaluation of the applicability of the developed method to the determination of the UO_2 distribution and total U^{235} load in UO_2–Zircaloy-2 compacts. However, the method will be equally applicable to the finished elements, requiring only recalibration. Although the method is rather generally applicable to enriched fuel plates, the calibration data described is directly applicable only to UO_2–Zircaloy-2 compacts of the following general specifications:

Length	3 to 6 in.
Width	0.62 in.
Thickness	0.08 in.
U^{235} concentration	0.44 to 0.86 g/in.
UO_2 particle size	$\sim 4 \times 10^{-3}$ in. dia.
Nb coating on UO_2	0 to 3×10^{-5} in.

SUMMARY

The horizontal homogeneity of UO_2–Zircaloy-2 dispersion type fuel specimens can be determined with a precision of slightly better than ±5 per cent at the 90 per cent confidence level using 1-min counts over ¼-in. increments of fuel length. The present specification calls for homogeneity of ±5 per cent over 1-in. increments. Total fuel load in the defined compacts can be determined to a precision of at least ±1 per cent at the 90 per cent confidence level using 1-min counts over fourteen ¼-in. increments of fuel length. The method is, in general, applicable to any plate with fuel loads in the range 0.2 to 1.0 grams U^{235} per inch and with fuel thickness below 0.2 in.

PRINCIPLE OF THE METHOD

Uranium-235 emits a number of gamma photons in association with its primary alpha decay mode. The most abundant photon emitted possesses an energy of about 0.19 Mev. The detection of this photon presents a useful method for the determination of U^{235} in a mixture of uranium isotopes since none of the other naturally occurring isotopes of uranium, or their daughters, emit photons in this energy region. Another U^{235} photon at 0.095 Mev might be employed in this method but uranium K X-rays and photons from U^{234}, Th^{231}, and Th^{234} are also found in this energy region, complicating the interpretation of observed 0.095 Mev photon intensities. Using the 0.19 Mev photon intensity as a measure of the quantity of U^{235} presented to a photon detector, it is possible to scan an element along its length, through a suitable collimating arrangement, and measure the variation in U^{235} concentration. The material of which the viewing slit or collimator is constructed must present a large absorption cross section to the 0.19 Mev photons in order that the detector "see" only those photons emitted from that area of the fuel defined by the viewing slit. Lead is the obvious choice for collimator material. The mass absorption coefficient of Pb for 0.2 Mev photons is 0.942 cm^2/g. Using 11.3 g/cm^3 as the density of Pb, a $\frac{3}{8}$-in. thick Pb absorber will transmit only 0.0045 per cent of the incident 0.2 Mev photons.

Since uranium and zirconium both present significantly large absorption cross sections to 0.2 Mev photons, the intensity vs. fuel thickness function is not linear. For this reason, the method described is applicable only to fuel plates with cores less than about 0.2 in. thick. Above this thickness the sensitivity—change in observed intensity per unit change in U^{235} concentration—falls off rapidly as a function of fuel thickness.

A number of conventions can be used in expressing the distribution of U^{235} within a fuel plate. Since the method used here measures the 0.19 Mev photon intensity in a slit viewing the full plate width and scanned incrementally along the fuel length, homogeneity is expressed in terms of U^{235} weight per unit fuel length (U^{235} g/in.). The method will not detect inhomogeneity within the slit width nor will it detect symmetric inhomogeneity across the fuel plate width or inhomogeneity in depth.

SCANNING TECHNIQUE

The fuel compacts are scanned across the full width in $\frac{1}{4}$-in. increments along their length using a lead absorber $\frac{3}{8}$ in. thick containing a slit $\frac{1}{4}$ in. wide by 1 in. long. The fuel element rides on a spring loaded carriage which maintains the upper surface of the element in direct contact with a thin polyethylene sheet covering the lower surface of the absorber. The fuel element is moved under the slit in $\frac{1}{4}$-in. steps by a manually operated screw arrangement attached to the carriage. Positive position measurement is obtained by a 0.01 in. revolution odometer attached to the drive screw.

DETECTOR ARRANGEMENT

The detector used is a NaI (Tl) crystal $1\frac{1}{2}$ in. in dia. by 2 in. high centered on the slit at the upper surface of the lead absorber. The light pulses produced in the crystal by all photon interactions are "seen" by a DuMont 6292 multiplier phototube in which they are converted to electron pulses, amplified and subsequently sent to an Atomic Model 219A Preamp and a Baird-Atomic Model 510 single channel, differential pulse height analyzer. As the name implies, the differential pulse height analyzer provides a method of viewing the amplifier output voltage pulses incrementally and thus permits the determination of the number of pulses occurring in any amplitude increment and therefore in any increment of photon energy. The overall gain of the system is adjusted to 0.01 Mev/volt, i.e., a 1 Mev photon interaction appears as a 100-volt pulse at the amplifier output. The linearity of the system is checked using sources emitting known photon energies. For example, Fig. 1 shows the spectrum obtained by mounting a Cs^{134} source on the carriage in place of the fuel element. The 0.8 Mev and 0.605 Mev photons emitted during the decay of this isotope are seen as photopeaks at 80 and 60.5 volts using the differential pulse height analyzer. A plot of pulse height vs. energy shows the system to be linear down thru zero energy (Fig. 2). The NaI (Tl) crystal used here is much larger than required for efficient detection of the 0.19 Mev photons emitted by U^{235} (over 99.9 per cent of these photons are absorbed in 2 in. of NaI). The large crystal is employed to optimize the resolution of the photon energy of interest.

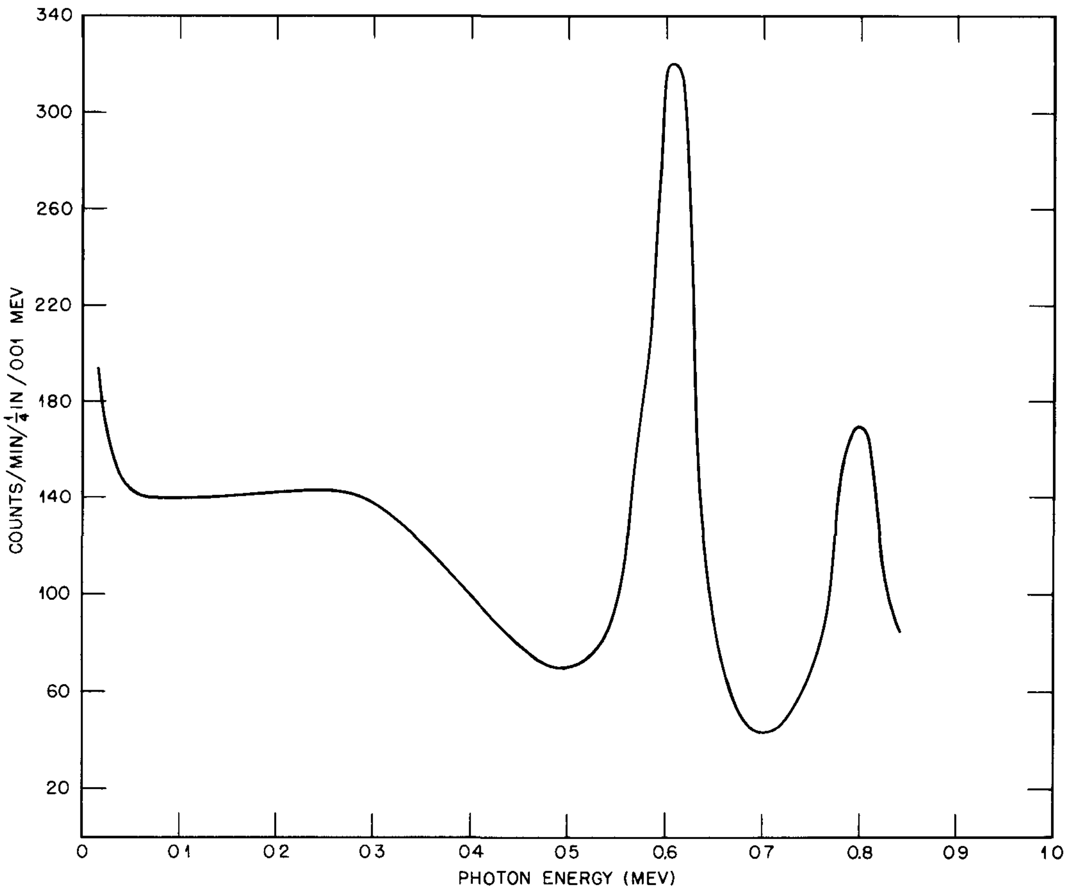

Fig. 1 — Cesium-134 spectrum, 0.01 Mev per volt. Counts/min $\Sigma_{0.5}^{\infty}$, 17,404 ± 129 counts/min (95 percent, 4 min); background, $\Sigma_{0.5}^{\infty}$, 74 counts/min.

RECORDING SYSTEM

The pulses falling within the selected increment of voltage (photon energy) at the pulse height analyzer are sent to a Baird-Atomic Model 988A Decade Scaler where the numbers detected in any selected period of time are manually counted. Other recording systems can be used, e.g., integrating count-rate meter, current measurement, and strip chart recording, but the manual counting technique over a selected time interval is the best method of obtaining optimum counting precision.

FUEL PLATE GAMMA SPECTRUM

A UO_2–Zircaloy-2 dispersion type fuel compact containing nominally 15 vol. % UO_2 shows the gamma spectrum indicated in Fig. 3 using the instrument described above. The photopeak at about 0.2 Mev is the peak utilized in this method of U^{235} distribution determination. The electronic window width of the differential pulse height analyzer is set at 5.0 volts. The lower discriminator is set at 17 volts so that all pulses between 17 and 22 volts are counted, i.e., all photon interactions dissipating an energy between 0.17 and 0.22 Mev in the crystal are counted. The shaded area under the spectrum of Fig. 3 represents the area of interest in this analysis. A wide increment of energy is utilized rather than a narrow increment around the center of the photopeak in order to minimize the effect of small variations in over-all gain on observed counting rates.

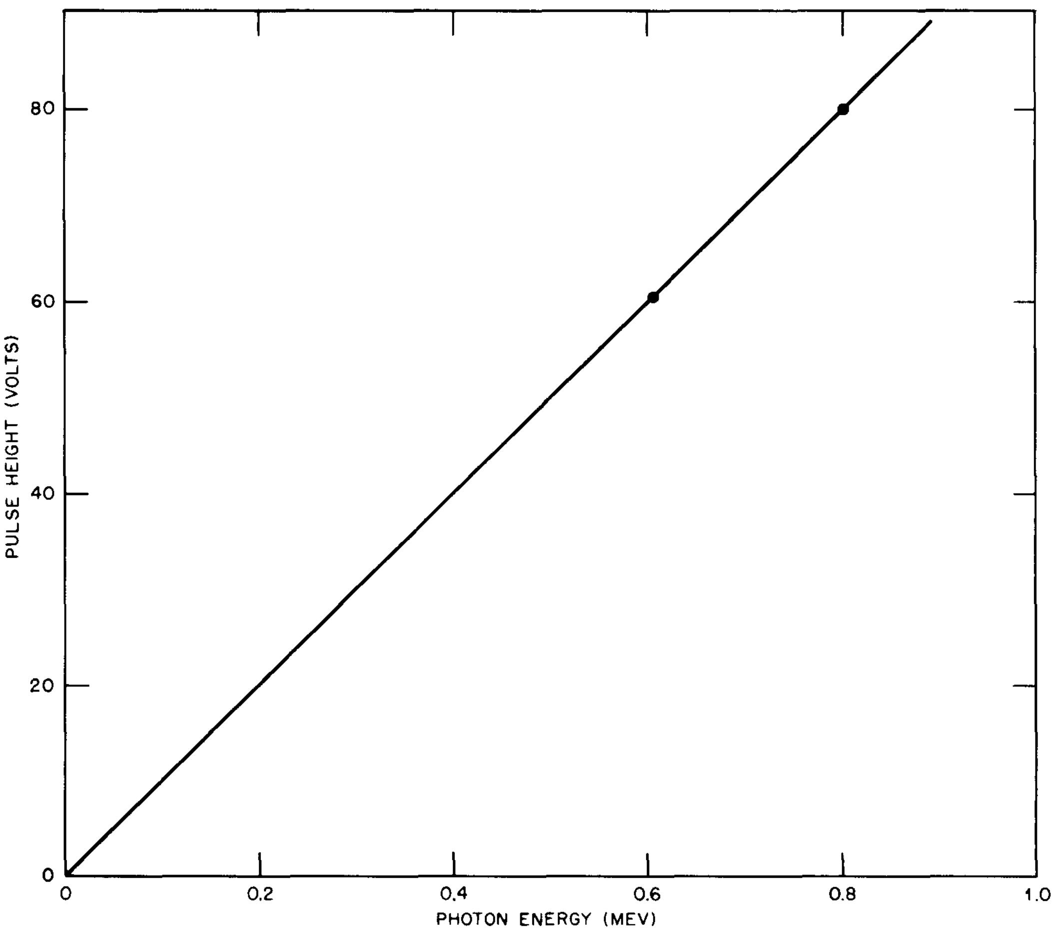

Fig. 2—Amplifier output pulse height vs. photon energy.

SENSITIVITY OF METHOD

The sensitivity of the method, change in observed counting rate per unit change in U^{235} content per unit fuel length, was determined by integrating and averaging the counting rate observed in ¼-in. increments over the entire length of 2 UO_2–Zircaloy-2 compacts containing measured quantities of U^{235}. Fourteen increments were counted for 2 min each on UO_2–Zircaloy-2 compacts containing 0.440 and 0.866 grams of U^{235} per inch, nominally 7.5 vol. % UO_2 and 15 vol. % UO_2, respectively. The integrated rates were 186,071 counts/min and 336,657 counts/min with precisions of ±0.3 per cent and 0.2 per cent at the 95 per cent confidence level. Therefore, the average counting rate per ¼-in. increment for each of these compacts was 13,291 and 24,047 counts/min/¼ in. with the same per cent precisions. Figure 4 shows a plot of this counting rate as a function of U^{235} concentration. The sensitivity, indicated by the slope of the line in Fig. 4, is 252 counts/min/0.01 g/in. At a counting rate of 24,000 counts/min, a 1-min count yields a precision of 1 per cent in the determined rate or ±240 counts/min at the 90 per cent level of confidence. The ±240 counts/min corresponds to an uncertainty in U^{235} content of ±0.02 g/in. of plate. The present specification for UO_2–Zircaloy-2 compacts (as indicated in drawing KAPL 953B919) requires that the U^{235} content per inch of plate not vary from the average concentration by more than ±5 per cent or, in the 0.866 g/in. case, by more than ±0.022 g/in. Since the method used here measures the U^{235} content in units of ¼ in. along the plate surface rather than in 1-in. increments, it is evident that this method presents a more than adequate means of monitoring UO_2–Zircaloy-2 compacts for specification homogeneity. At the lower uranium loading, nominal 7.5 vol. % or 0.44 grams of

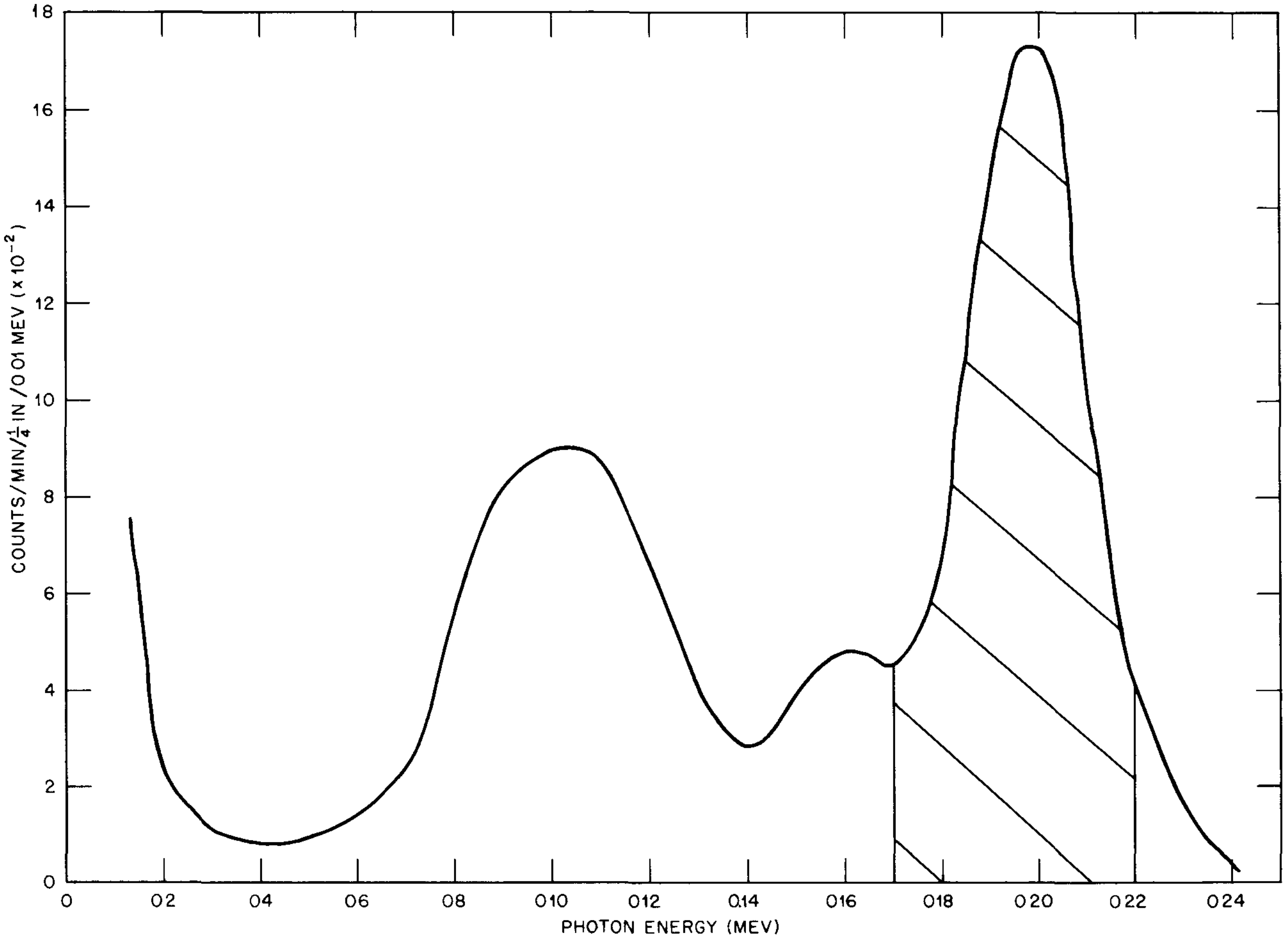

Fig. 3—Gamma spectrum, 15 vol. % UO_2 compact (gain, 0.01 Mev per volt).

U^{235} per inch, the 13,791 counts/min rate is determined to a precision of ±1.5 per cent at the 90 per cent limit or ±200 counts/min.

UO_2-ZIRCALOY-2 COMPACT SCANS

Determination of the distribution of U^{235} in three separate UO_2-Zircaloy-2 compacts are shown in Figs. 5 and 6. These figures are essentially self-explanatory. Each point represents a 1-min count over the ¼-in. increment. Compact No. Z-50 represents a compact fabricated using a purposely inadequate mixing technique. The two compacts shown in Fig. 6 were fabricated using the best available technique. The compacts of Fig. 6 fail to meet the specification of ±5 per cent per inch if based on the ¼-in. increments. However, they meet the specification if the average concentration over a 1-in. increment is accepted.

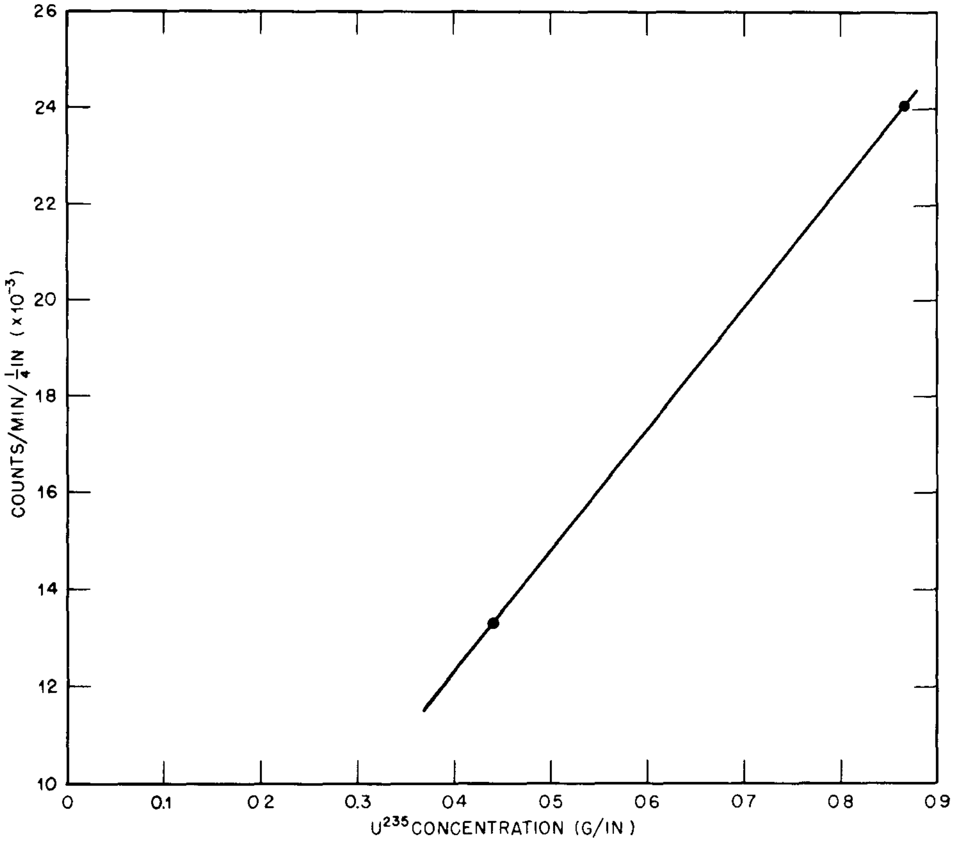

Fig. 4—Counting rate, $\Sigma_{0.17\text{ Mev}}^{0.22\text{ Mev}}$ as a function of U^{235} concentration (0.08 in. thick by 0.62 in. wide UO_2-Zircaloy-2 compacts; slope, 252 $\frac{\Delta\text{counts/min}}{\Delta 0.019/\text{in.}}$).

DETERMINATION OF TOTAL U-235 LOAD/ELEMENT

The integrated counting rates over 14¼-in. increments indicated on Fig. 6 represent a measure of the total fuel load of these plates. Accepting these plates as standards, they represent a method of determining total fuel load of all subsequent plates. The precision indicated is based on 2-min counts per increment. Since it is expected that only 1-min counts per increment will be used in routine analysis, the total load is determinable to a precision of about ±1 per cent at the 90 per cent confidence level. Although all of the calibration data

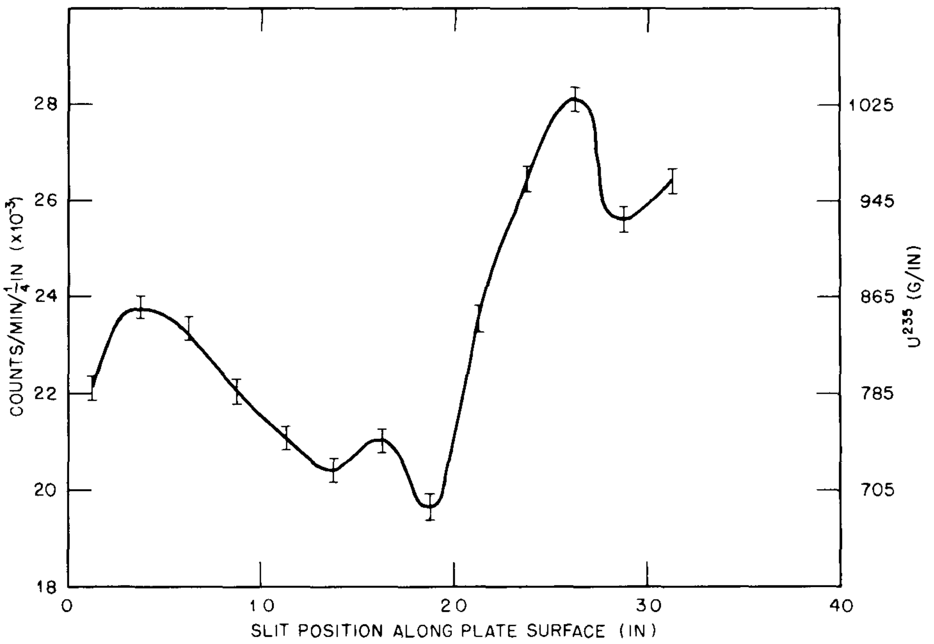

Fig. 5 — Homogeneity determination, compact No. Z-50 (15 vol. % UO_2). This compact was prepared using a purposely inadequate mixing technique.

quoted here are based on accepting the total load of these two reference plates based on fabrication weights, these plates are now being destructively tested to confirm the values used.

NONRANDOM ERRORS

In all of the above quotations of precision, the basic assumption is made that only random counting error is acting to influence the precision of the method. Nonrandom errors might be introduced by variations in several of the system parameters: phototube gain, pre-amp gain, amplifier gain, lower discriminator bias, upper discriminator bias, high voltage, count time, and fuel to detector geometry. The system has been designed to eliminate or minimize each of these error sources. That, indeed, they have been satisfactorily eliminated is confirmed by the facts that a day-to-day gain check reveals that the pulse spectrum is shifted less than 1 volt/day and that a chi-square test for comparing theoretical and observed variance shows an insignificant value, $X^2 = Ns^2/\sigma^2$, for the variance of 10 1-min counts of the 0.2 Mev U^{235} photopeak.

Repositioning of a single element seems to offer the greatest source of imprecision. However, since extremely exact location of inhomogeneity is not a variable of large importance, the repositioning variable is not critical. A series of counts of the 0.2 Mev photopeak involving repositioning between each count showed a chi-square value in the zone of questionable significance, i.e., significant at the 95 per cent but not significant at the 99 per cent level of reliability. Some modifications of the scanning mechanism now in progress may clarify this relatively minor problem.

Another nonrandom error is introduced by a small "end effect" in the fuel scanning method. An 0.866 g/in. compact placed with its leading edge 0.05 in. from the near edge of the slit produces a counting rate of 230 counts/min in the 0.17 to 0.22 Mev region. This is about 10 per cent of the analytical rate for this compact. With the leading edge located just at the near edge of the slit a rate of 1900 counts/min is observed. In both cases the slit is shielded with a 0.08-in. thick Zr plate as a stand-in for the fuel compact. This observation has significance in two respects. First, the ¼-in. increment is not exactly defined, i.e., 0.05 in. of the compact on each side of the slit contributes significantly to the determined counting rate but, of course, not in proportion to equal increments of the plate surface located directly below the slit. The net effect is a very slight broadening of the defined slit. Second, when a determination is made with the slit located at the last ¼ in. of the compact, the observed count rate will be reduced by the number of counts contributed by the 0.05 in. of fuel normally located immediately adjacent to one side of the slit. If a rate is determined for this last ¼-in. increment of the plate, it must be corrected upward by about 1500 to 1900 counts/min for the nominal 15 vol. % compacts. In determining total fuel load the last 0.05 in. on each end of the plate is ignored. This end effect is ignored, also, in the scans of Figs. 5 and 6 where the first point on the left of each scan represents the first ¼ in. of the plate.

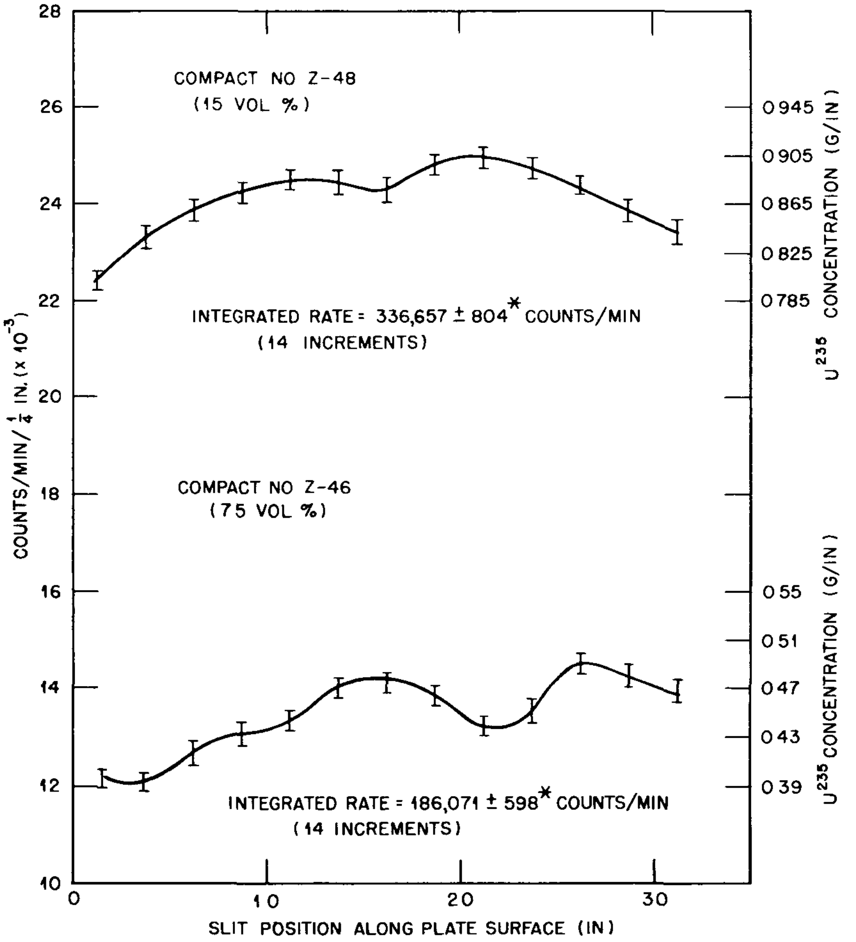

Fig. 6—Homogeneity and total load determination. (*Precision of summation of rate observed over 14¼-in. increments using 2-min counting time per increment, 95 per cent level of reliability.)

THE DETERMINATION OF INHOMOGENEITY IN DEPTH

It may be possible to determine the U^{235} distribution in depth within the ¼-in. increment by measuring the ratio of the 0.19 Mev peak to the 0.1 Mev peak (see Fig. 3). Some preliminary measurements have been completed in an attempt to evaluate this method, with some success. However, the principal limitation appears to be the necessity of obtaining rather precise evaluation of each of the rates involved so that the resulting ratio can be defined with a precision sufficient to provide reasonable sensitivity to ratio changes. Counting times of the order of 20 to 30 min may be required, placing a definite restriction on the application of the technique in routine use. In addition, various symmetrical distributions within the increment scanned will negate the applicability and large horizontal inhomogeneities will reduce the sensitivity of the ratio method.

ROUTINE ANALYSIS OF ENRICHED URANIUM WITH A GAMMA PULSE HEIGHT ANALYZER

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ABSTRACT

The uranium content of solutions and solids obtained during salvage operations of enriched uranium is measured for both process control and inventory. Measurements are made with a single channel differential pulse height analyzer. The analyzer is driven by the gamma radiation detected in a well type scintillation counter. Pulse height analyses have a variation of 1 per cent on nitrate solutions containing 500 ppm to 16 per cent by weight enriched uranium. Some discard level material cannot be measured by this method because of the interference of other gamma emitters. Analysis time has been reduced to one-fourth of the time previously required by color or volumetric methods. Graphites and filter muds are analyzed directly, whereas, previously a tedious separation was required before a chemical determination could be made.

INTRODUCTION

In normal manufacturing operations, if scrap cannot be easily reprocessed, it is either sold or discarded. With enriched uranium every particle must be salvaged or accounted for. The salvage department recovers enriched uranium from solids such as crucibles, molds, wiping tissues, and from organic liquids such as cooling and lubricating oils. In the processing of these wastes the amount of enriched uranium must be known for criticality reasons to handle or store it within safe dimensions. An SS accountability inventory is taken each month. For these reasons a variety of samples are sent to the Analytical Laboratory for the determination of enriched uranium. From 200 to 600 samples are received a month. Speed and reasonable accuracy are required.

EQUIPMENT

The instrument used for this work is a Nuclear Chicago Model 1820 Recording Spectrometer, which is a single channel differential pulse height analyzer that automatically scans the pulse height spectrum and graphically records the input pulse rate vs. pulse height. (Fig. 1) For routine work, the recorder is not used, but instead, the pulses are counted on a Technical Measurement Corporation Model SG 3A 2 Scaler. In addition to the recording spectrometer, a Nuclear Chicago Model 132 Analyzer Computer is used. (Fig. 2) A Nuclear Chicago Model D 85 Scintillation Detector is also used with both instruments.

We are expecting delivery on a Packard Instrument Co. Model 400-A, and 400-B automatic sample handling device with automatic readout. This equipment will allow us to load up to 100 samples at a time and to operate it 24 hours each day without attendance other than during the loading period.

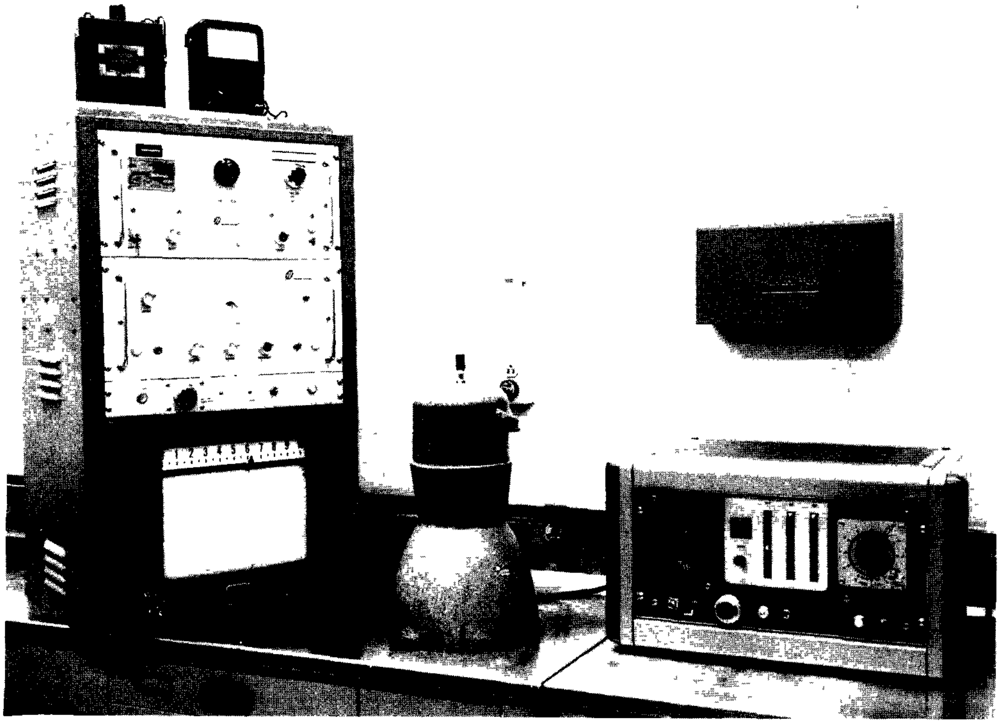

Fig. 1—Nuclear Chicago Model 1820 recording spectrometer.

Fig. 2—Nuclear Chicago Model 132 analyzer computer.

THEORY OF OPERATIONS

In the measurement of radioactive nuclides which emit gamma rays, the use of a gamma ray spectrometer is comparable to the use of an ordinary spectrograph in analytical chemistry. A gamma ray spectrometer is also called a gamma pulse height analyzer. The gamma radiation emitted by a radioactive nuclide is electromagnetic radiation identical in fundamental nature with visible, infrared, and ultraviolet light found in ordinary spectroscopy.

Table 1 gives a comparison of the energy and wave length with the more familiar electromagnetic radiations.[1] The individual photons can be absorbed in a crystal of sodium iodide containing traces of thallium. A single gamma photon is converted to a large number of photons of visible light which can be measured with a photomultiplier tube. Since the number of visible photons is proportional to the gamma ray energy, the energy of the gamma ray photon can be determined.

Table 1—COMPARISON OF ENERGY AND WAVELENGTH

Radiation	Energy per photon, ev	Wavelength, A
Infrared	1.5	8,000
Visible (green)	2.25	5,500
Ultraviolet	3.1	4,000
Nuclear gamma rays	$1 \times 10^4 - 1 \times 10^7$	0.001 – 1.2

A gamma ray spectrometer is made up of a suitable scintillator (i.e., NaI crystal with traces of Tl) optically coupled to a photomultiplier tube, a highly regulated power supply, a linear amplifier, a single channel differential analyzer, and a scaling circuit. The pulses from the photomultiplier are rather small and must be amplified. An amplifier with good linear response is necessary to keep the proportionality between the energy of the photons and the amplitude of the pulses.

After amplification, a selection is made of the pulses lying within a narrow range of pulse heights. This is done by a differential analyzer, which observes all the pulses, but activates the scaler, or a rate meter recorder, only when the incoming pulse is within a previously selected range. The analyzer contains a lower and upper discriminator. If the pulse height is lower than the lower level discriminator it is discarded. If the pulse height is large enough to exceed the settings of both the upper and lower discriminators, the pulse is rejected by an anticoincidence circuit connecting the outputs of the two discriminators. Only if the pulse is large enough to exceed the lower level, but not so large as to reach the upper level is an output pulse produced. Figure 3 shows how the differential analyzer picks out the wanted voltage.[2] The output pulses correspond to a given rate of energy and are measured on a scaler or a rate meter with a recorder. When a scintillating crystal coupled to a photomultiplier is exposed to gamma radiation of a given energy such as U^{235} 184 kev, one obtains a pulse distribution as shown in Fig. 4.

PROCEDURE

1. Pipette a 3 ml aliquot into a 15 by 125 mm test tube. The aliquot should not contain too much enriched uranium because too high a count will cause the analyzer to malfunction. It is best to keep the counting rate less than 3×10^6 counts/min. We do not operate much over 1.5×10^6 counts/min.
2. Count for 6 min.
3. Calculate, using the standard values obtained just above and just below the unknown. A set of standard tubes, each containing 3 ml, is prepared by dilution of a solution of uranium metal of known isotopic concentration. The series desired cover the range of 0.005 to 150 g/liter in steps; i.e., 150, 100, 50, 25, 10, 5, 2, 1. The metal is dissolved in nitric acid, evaporated to dryness and made to volume with 5 N HNO_3. All dilutions are made with 5 N HNO_3.

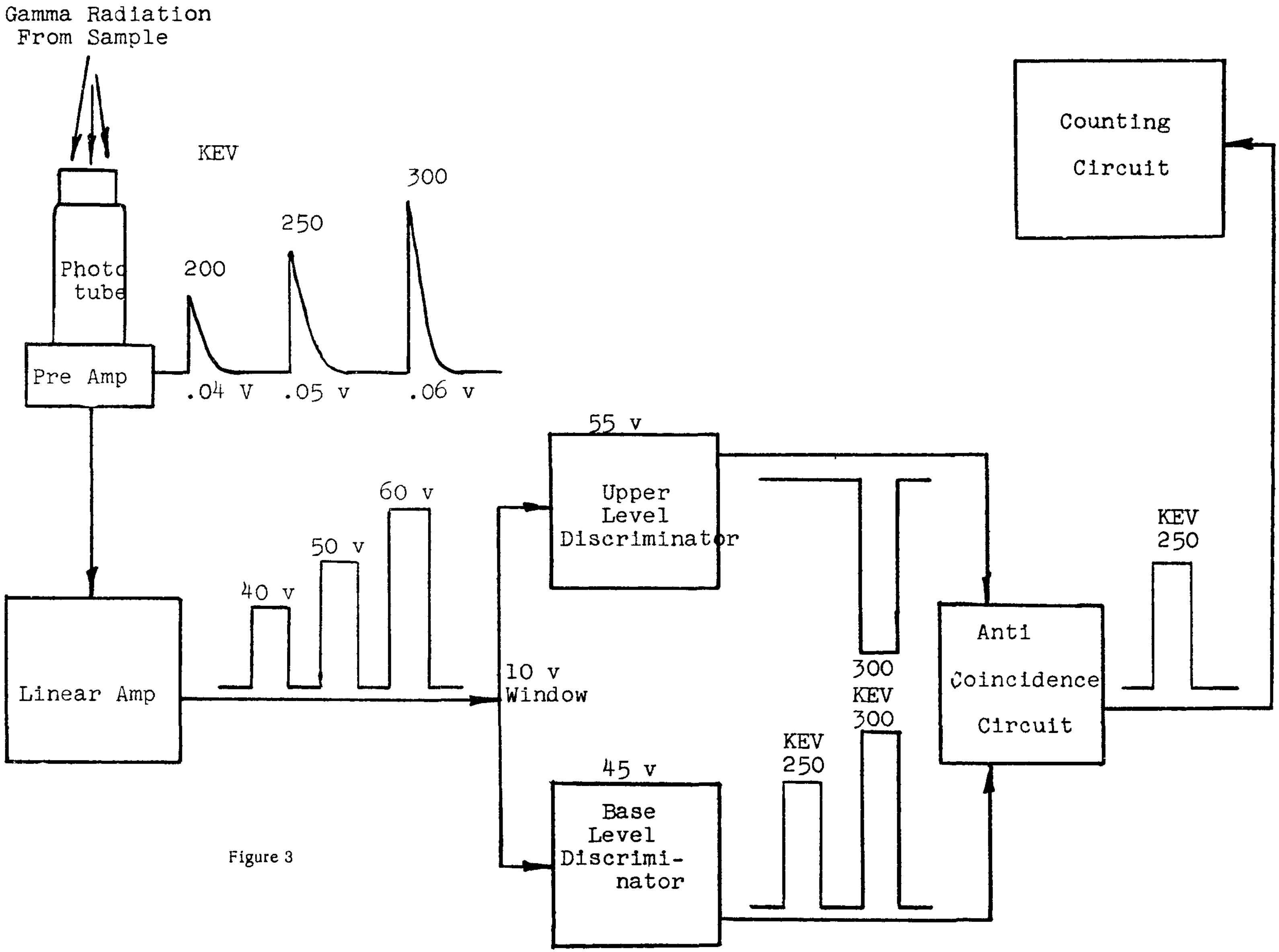
Gamma Radiation
From Sample
Photo
tube
Pre Amp
KEV
200
250
300
.04 V
.05 v
.06 v
Linear Amp
40 v
50 v
60 v
10 v
Window
55 v
Upper
Level
Discriminator
45 v
Base
Level
Discrimi-
nator
300
KEV
300
KEV
250
Anti
Coincidence
Circuit
KEV
250
Counting
Circuit

Figure 3

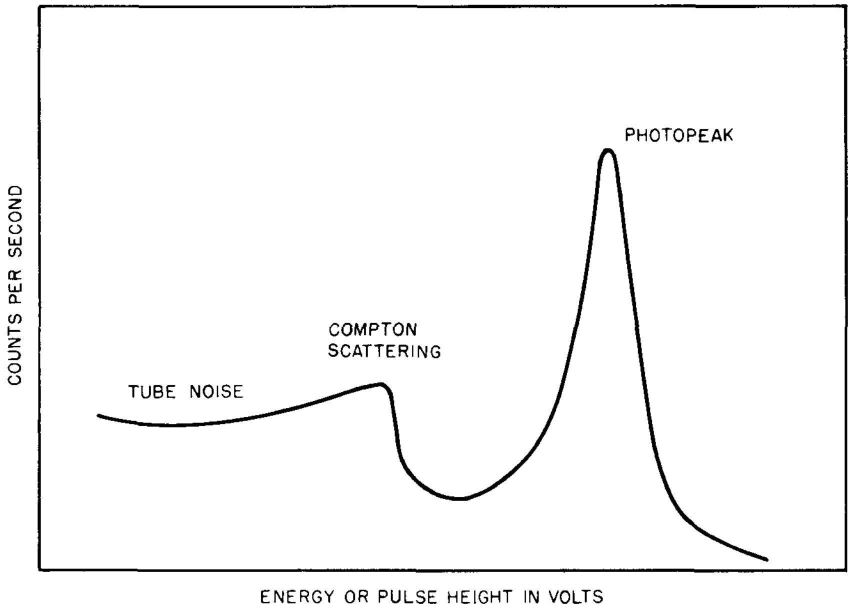

Figure 4

Calculation (Fig. 5):

$$\frac{(C_{s^1} - C_{s^2})\ (D_x - D_{s^2})}{(D_{s^1} - D_{s^2})} + C_{s^2} = C_x$$

where C_{s^1} = concentration of a standard higher than the unknown.
C_{s^2} = concentration of a standard lower than the unknown.
D_{s^1} = counts in 6 min of high standard.
D_{s^2} = counts in 6 min of low standard.
D_x = counts in 6 min of unknown.
C_x = concentration of unknown.

All units must be kept consistent. If $C_s{}^1$ and $C_s{}^2$ are in grams per liter, C_x will be also in grams per liter. If D_x is total counts in 6 min, the $D_s{}^1$ and $D_s{}^2$ must also be total counts of these respective standards in 6 min. If a gram per gram answer is desired, the density of the sample must be determined and appropriate correction made in the calculation.

$$\frac{\text{grams per liter}}{\text{Density}} \times 10^{-3} = \text{g/g}$$

A calibration curve (Fig. 6) of counts per 6 min vs grams per liter is not used for calculations because when the method was first set up the spectrometer drifted so much from hour to hour that consistent results were only obtained by bracketing the sample. At the present time this method of calculation is still used, but the standard values are used without bracketing because a standard is run every hour and if necessary the analyzer is adjusted to give the standard value.

When filter muds are analyzed a spike technique is used. That is, a known amount of enriched uranium is added to the sample, and the sample count is corrected for the effect the sample has on the count of the known amount of uranium. Any samples which are of unusual composition are spiked and corrected if necessary. The quantity of enriched uranium oxide can be determined in graphite. The samples are ignited at 850°C for a half hour to convert any uranium metal to U_3O_8. The samples are checked for ignition loss, but graphite does not oxidize readily. The samples are ground with a mortar and pestle, and a 3 ml volume measured into a tared test tube and weighed. The graphite samples vary in density so that a constant sample weight changes the sample volume enough to effect the counting geometry. Standards are prepared by weighing mixed known amounts of graphite and U_3O_8. See Fig. 7.

CHEMISTRY LABORATORY WORK CARD GAMMA

G

T ____________________ Samp. Wt.

N

Cs_1: conc. of std > sample (g/l)
Cs_2: conc. of std < sample (g/l)
Ds_1: disintegration rate of S_1
Ds_2: disintegration rate of S_2
Dx : disintegration rate of sample

Aliquot size ____________________ Dilution ____________________

(Cs_1 ________ $-Cs_2$ ________)(Dx ________ $-Ds_2$ ________) = ____________
(Ds_1 ________ $-Ds_2$ ________) +Cs_2 ____________

____________ g/l

(____________ g/l)(Dil. Fact. ____________) = ____________ g/g
(Samp. Wt. ____________)(10^3)

(____________ g/l)(10^3)(Dil. Fact. ____________) = ____________ ppm
Samp. Wt. ____________

By ____________ Date ____________ No. Detns. ____________ Answer ____________

Req. No.

Type

Date

Chg.

Cont. No.

Sp. Gr.

Net Wt.

Total U

g/g ☐
g/l ☐
ppm ☐

Figure 5

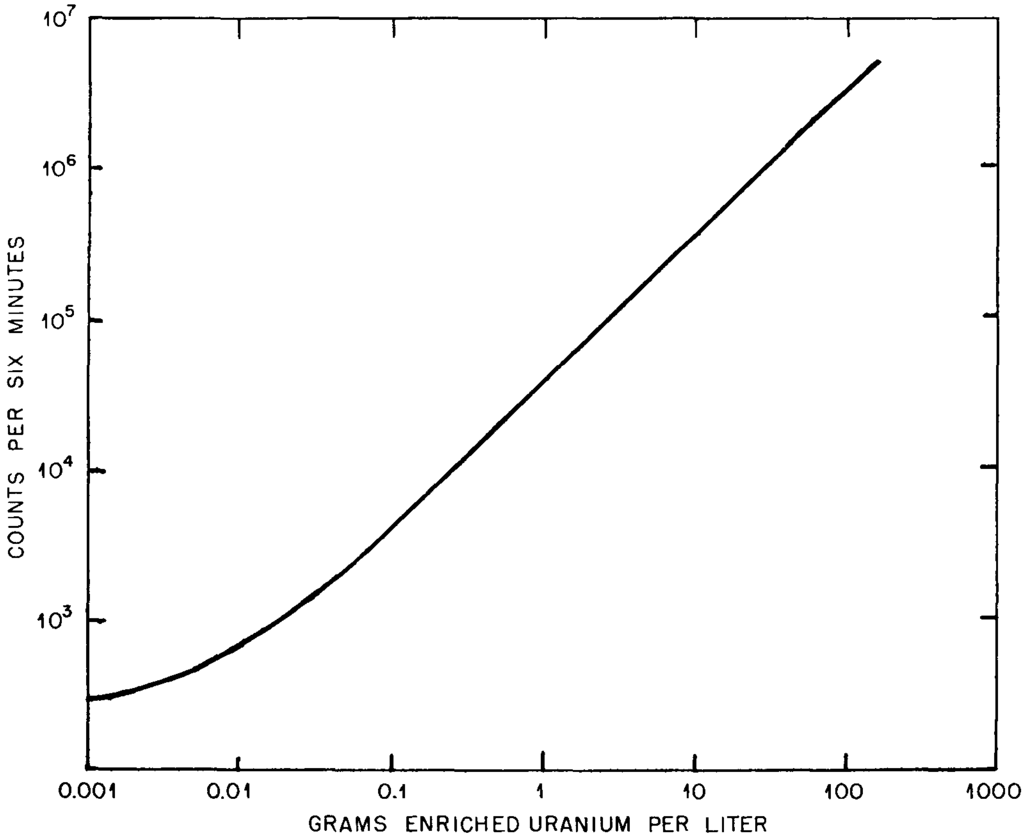

Fig. 6—Counts per six minutes vs grams enriched uranium per liter.

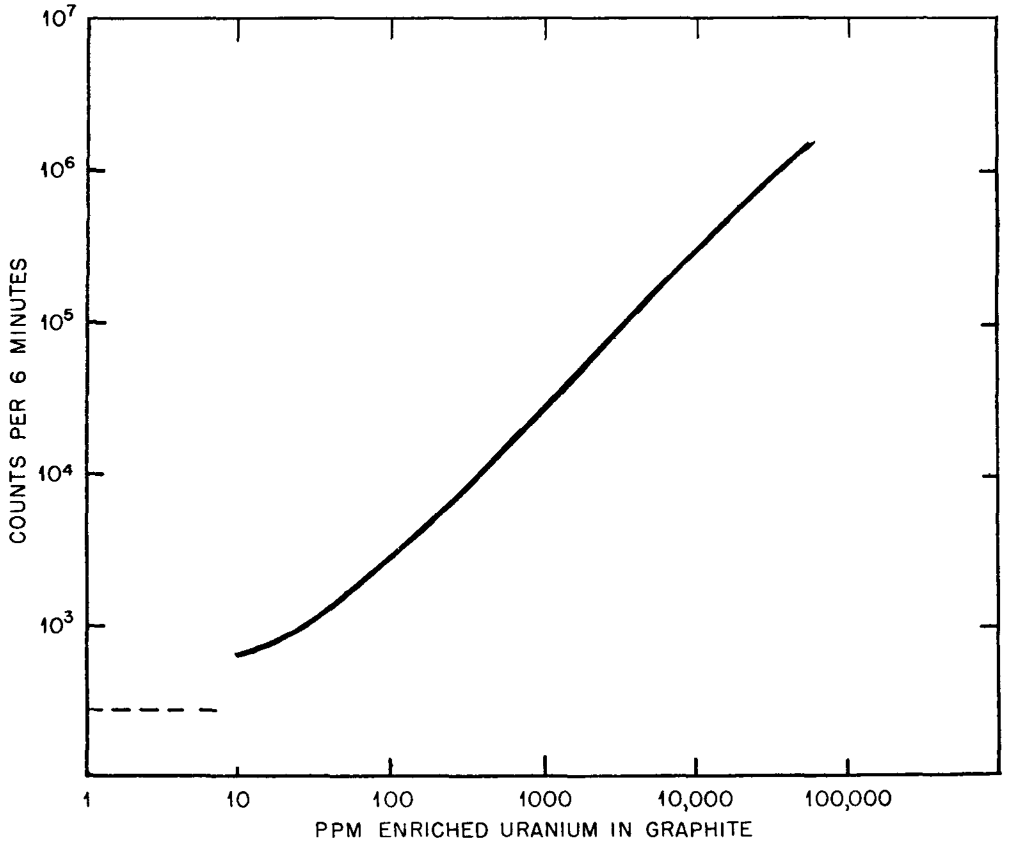

Fig. 7—Counts per six minutes vs ppm enriched uranium in graphite.

SAMPLE SIZE

A 3 ml sample aliquot was chosen for counting for two reasons. Most of the samples of the enrichment we handle, have sufficient count that 3 ml is enough sample. Some samples have so much count they have to be diluted to a definite volume and a 3 ml aliquot taken. Counting efficiency is better with a 3 ml sample for this particular crystal and the diameter of the test tube containing the sample has less effect on the count. Most of our analyses must be reported on a volume basis. To obtain the greatest accuracy, 3 ml Normax pipettes are used. They were found to deliver within ±0.23 per cent of 3 ml. The test tubes 15 by 125 mm varied in diameter from 14.9 to 15.6 mm. This much variation can cause a difference of 0.8 per cent in duplicate analysis with a 3 ml sample. Results could vary as much as 1.4 per cent with a 5 ml sample. To eliminate this variable, the tubes were sized with a micrometer to fall within 0.590 in. to 0.595 in. diameter. This reduces the effect of tube diameter to less than pipetting errors.

Figure 8 shows the effect of volume on counting efficiency. A 1 ml sample having a count of 338,300 counts/6 min was successively diluted with 1 ml portions of distilled water. The amount of U^{235} was kept constant while the volume was increased. At 10 ml volume, the counting efficiency was only 53 per cent of that obtained with a 1 ml volume.

Figure 9 shows the effect of sample volume vs count. One ml aliquots of the same solution were successively added to a test tube and counted after each addition. The U^{235} concentration was increased in direct proportion to the sample volume. After 10 ml volume, the count becomes relatively constant so that measurement of 10 ml would not be critical, however, the tube diameter would be critical. To use 10 ml sample size in our work would require that most of the samples be diluted, i.e., made to volume and a 10 ml aliquot taken.

INSTRUMENTATION

Pulse Height Analyzer Control Settings

Window width	2 volts	10 volts
Photomultiplier	730 volts	730 volts
Base Level	27 volts	23 volts
Gain Setting	16	16
Resolution	25%	55%
Energy Range	173–194 kev	131–237 kev

Instrument Log

1. Date and time of each entry.
2. Standard value counted.
3. Six minute count.
4. Percent variation from norm.
5. High voltage setting.
6. Base level setting.

The variation from the norm usually holds within ±0.2 per cent when the 10 g/liter standard is counted. If the total count is too low decrease the high voltage adjustment by one volt increments. If the total count is too high, increase the high voltage. The base level may need adjustment at the same time to assure that the window is on peak. To peak in the instrument turn the base level 5 volts above the standard setting and make a 1 min count. Then set the base level at 5 volts below the standard setting and make a 1 min count. The difference between the standard setting and the high and low settings should be the same. If not, adjust the base level until they are the same.

Any variation in high voltage applied to the photomultiplier affects the count rate. Erratic results or shifting of the peak is usually associated with a change in the voltage. To assure that this voltage is constant an arrangement was made as shown in Fig. 10 with battery and vacuum tube voltmeter. The battery is a WS Eveready high voltage battery set on taps 2 and 7 to deliver 765 volts. The Simpson vacuum tube voltmeter is set on the 12-volt scale and reads

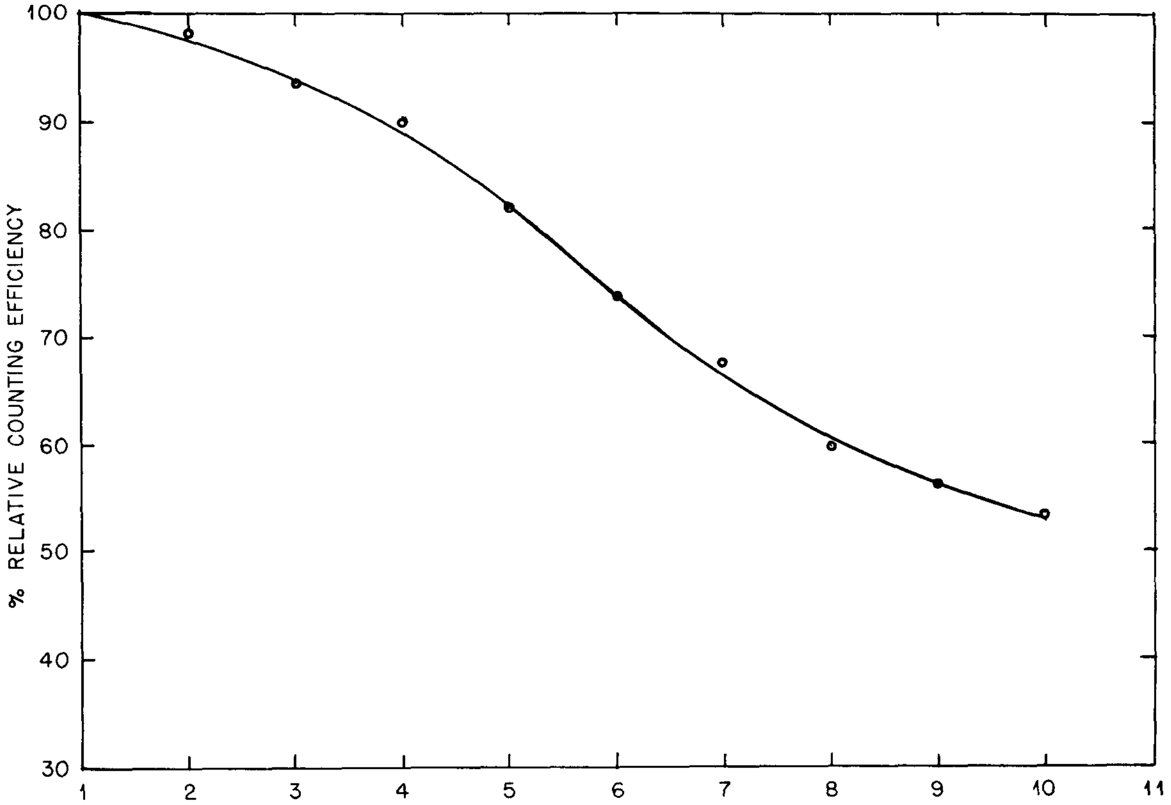

Fig. 8—Effect of volume on the count rate with a constant among of U^{235}.

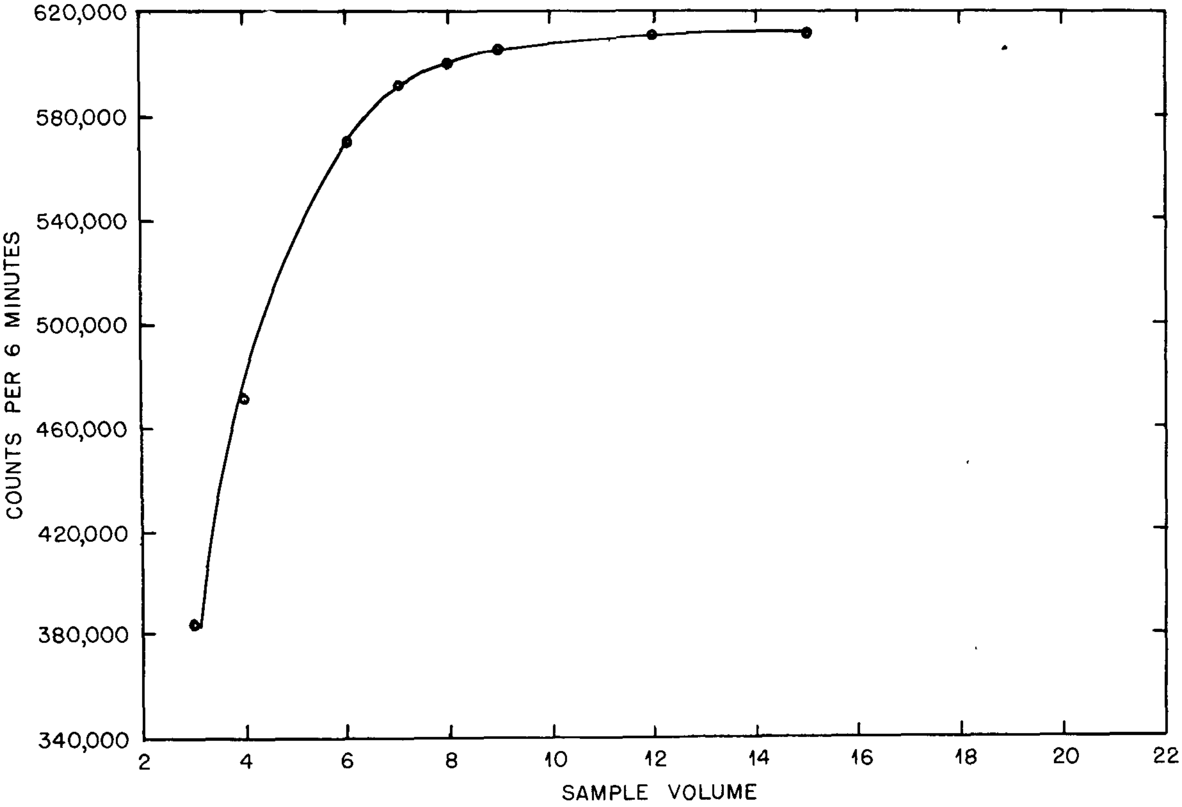

Fig. 9—Variation of count rate with sample volume.

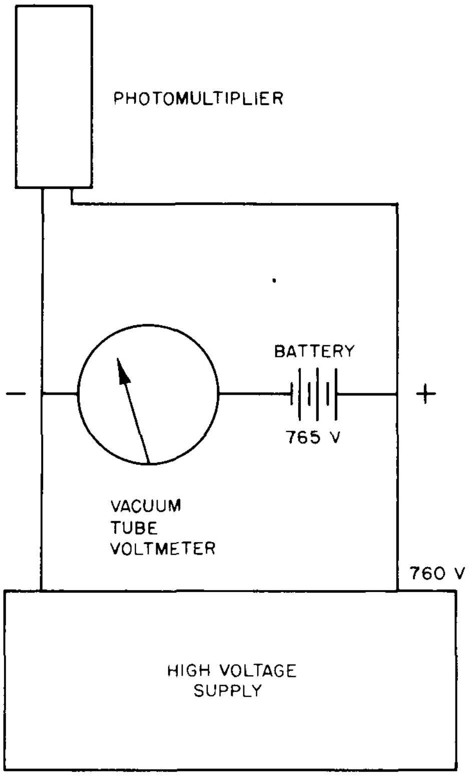

Figure 10

a differential voltage between the battery and the high voltage supply of around 2.5 volts. Any change in the high voltage supply will give some other reading than 2.5 volts. Meters reading directly to 760 volts were tried, but a change of 1 volt was impossible to read. The best signal to noise ratio and energy resolution are obtained from a scintillation counter when it is operated at low gain or with the minimum usable voltage applied to the photomultiplier tube.

COMPARISON DATA

The first work at Rocky Flats using a single channel pulse height analyzer for measuring enriched uranium was done by J. T. Byrne and M. Kreider[3] of our Analytical Methods Development Group. The reports on work done at the Hanford plant by U. L. Upson and D. G. Miller[4] as well as the work done at Mallinkrodt's St. Louis plant by S. D. Nelson and D. M. Bailey[5,6] were very helpful.

The first results on 103 production samples analyzed with pulse height analysis by M. Kreider when compared to the chemical results showed variations ranging from –13 to +12 per cent.

Table 2 shows a portion of this data for colorimetric analysis.

Table 3 shows a portion of this data for volumentric analysis.

Table 4 shows duplicate analysis for pulse height analysis.

Approximately 20 working hours were required to prepare and count these 103 samples by pulse height analysis. Approximately 200 hours were required to determine these samples by chemical analysis. Because this method offered such a savings in time, and effort, the February 1958 salvage inventory was calculated with this data.

Table 5 shows the inventory to have a bias of −0.07 per cent of the amount present analyzed by pulse height analysis and when compared to the inventory based on chemical analysis.

Table 2—COMPARISON OF COLORIMETRIC ANALYSIS WITH GAMMA COUNTING

Color g/liter	Gamma, g/liter	Difference,* %
0.0460	0.0477	+3.7
0.163	0.164	+0.6
0.363	0.357	−1.7
0.744	0.728	−2.2
1.57	1.49	−5.1
1.59	1.51	−5.0
1.69	1.65	−2.4
1.90	1.87	−1.6
2.00	1.92	−4.2
2.12	2.13	+0.5
2.55	2.52	−1.2
3.62	3.56	−1.7
4.63	4.78	+3.2
6.23	6.28	+0.8
13.3	13.2	−0.8
14.9	15.8	+6.0
22.5	22.0	−2.2
26.4	26.1	−1.1
61.7	59.7	−3.2

Average difference or bias, −0.9%

S.D. (Standard deviation) of differences, 2.9%

$$* \text{Difference } \% = \frac{\text{g/liter color} - \text{g/liter gamma}}{\text{g/liter color}} \times 100$$

Table 3—COMPARISON OF VOLUMETRIC ANALYSIS WITH GAMMA COUNTING

Volumetric, g/g	Gamma, g/g	Difference,* %
0.183	0.186	+1.6
0.240	0.245	+2.1
0.217	0.216	+1.7
0.213	0.217	−0.5
0.218	0.219	+1.9
0.218	0.219	+0.5
0.0436	0.0442	+1.4
0.157	0.160	+1.9
0.126	0.127	+0.8
0.0647	0.0645	−0.3
0.154	0.151	−1.9
0.114	0.113	−0.9
0.133	0.132	−0.8
0.123	0.123	0.0
0.112	0.112	0.0
0.119	0.120	+0.8

Average difference or bias +0.5%

S.D. of differences +1.2%

$$* \text{Difference } \% = \frac{\text{g/g volumetric} - \text{g/g gamma}}{\text{g/g volumetric}} \times 100$$

Table 4 — DUPLICATE ANALYSIS BY PULSE HEIGHT ANALYSIS

Concentration Range	Number Samples	S.D. of Differences
0.01 – 0.8 g/g	33	0.50%
8 – 300 g/liter	31	0.62%

Table 5 — COMPARISON OF CHEMICAL AND PULSE HEIGHT ANALYSIS FOR THE FEBRUARY 1958 INVENTORY

Concentration	Average difference, %*	Portion of total inventory, %
0.4 g/liter	5.5	0.05
1.8 g/liter	1.8	7.7
2.0 g/liter	0.0	0.7
6.0 g/liter	0.33	1.4
9.0 g/liter	−0.06	7.5
25.0 g/liter	−0.01	13.7
44.0 g/liter	0.27	6.5
70.0 g/liter	0.30	40.7
0.2 g/g	−0.31	22.0

Total inventory difference −0.07% of amount inventoried.
101 samples analyzed for the inventory.

$$* \text{Average difference in } \% = \frac{\text{chemical value} - \text{gamma value}}{\text{chemical value}} \times 100$$

Table 6 shows a bias of −0.04 per cent for the March 1958 salvage inventory covering 74 samples. The samples representing the lowest concentrations had the greatest differences but they represented only a small part of the inventory. Over 80 per cent of the amount of enriched uranium inventoried had a difference less than 0.5 per cent.

Table 7 shows a portion of the Measurement Control Report for December 1957 prepared by the Rocky Flats Accountability and Production Control Group. Samples for this data were submitted to the laboratory by the APC group either as control samples or disguised as production samples. These results were obtained by wet chemical methods.

Table 8 shows a similar measurement control report for February 1959. These results were mainly by pulse height analysis.

Table 9 shows a comparison of the control reports of December 1957 and February 1959. The pulse height results do not approach the variation obtained by wet chemical methods. They do have a variation of 1.2 per cent or less which represents the bulk of the material inventoried.

INTERFERENCES

All the samples analyzed were assumed to be at equilibrium with any daughter products[4] formed. Measurements made on enriched uranium freshly extracted with ether show some decrease in count, but the decrease in count was within the standard deviation of the method. Our enriched uranium samples must have a constant isotopic content of U^{235}. Some raffinates obtained in the purification of enriched uranium do contain a greater concentration of daughter products.[3] These raffinates contain less than 100 ppm enriched uranium and are determined by the fluorimetric method. A pulse height analysis of these raffinates gave results approximately five times too high. A gamma scan of all new samples prior to counting will detect interferences.

Table 10 shows the effect of the medium on the count rate. If the enriched uranium is present as uranyl nitrate in solution in either water or nitric acid, the count rate is not af-

Table 6—COMPARISON OF CHEMICAL AND PULSE HEIGHT ANALYSIS FOR MARCH 1958

Concentration	Average difference, %*	Portion of total inventory, %
1.6 g/liter	−3 1	7.6
9.7 g/liter	1.4	2.4
26.0 g/liter	2.7	4 4
34.0 g/liter	−1.2	20.5
46.0 g/liter	1.2	29.7
0.2 g/g	−0.3	34 4

Total inventory difference −0.04% of amount inventoried.

74 samples analyzed for the inventory

* Average difference in % $= \frac{\text{chemical value} - \text{gamma value}}{\text{chemical value}} \times 100$

Table 7—MEASUREMENT CONTROL REPORT DECEMBER 1957

Average standard value	Number of samples*	Method of analysis†	Bias	S.D.	Variation, %‡
0.14 ppm	22	F	0.0 ppm	0.05 ppm	36.0%
0.11 ppm	17	F	−0.2 ppm	0.03 ppm	27.0%
0.55 g/liter	26	C	−0.007 g/liter	0.009 g/liter	1.6%
1.1 g/liter	14	C	−0.035 g/liter	0.029 g/liter	2.6%
1.6 g/liter	39	C	−0.016 g/liter	0.025 g/liter	1.6%
39.0 g/liter	17	T	−0.07 g/liter	0.14 g/liter	0.36%
390.0 g/liter	11	T	0.00 g/liter	0.1 g/liter	0.02%
0.2 g/g	72	T	−0.0001 g/g	0.0007 g/g	0.36%

* Based on seven months period.

† F, fluorimetric analysis; C, colorimetric analysis; T, titration analysis.

‡ Variation % $\frac{\text{S.D.} \times 100}{\text{A.S.V. (average standard value)}}$

Table 8—MEASUREMENT CONTROL REPORT FEBRUARY 1959

Average standard value	Number of samples*	Method of analysis†	Bias	S.D.	Variation, %‡
2.4 ppm	17	F	−0.56 ppm	0.46 ppm	19
1.8 ppm	20	F	−0.32 ppm	0.61 ppm	33
34.0 ppm	12	G	1.2 ppm	2.2 ppm	6.4
0.62 g/liter	20	G	0.003 g/liter	0 008 g/liter	1.3
3.3 g/liter	17	G	0.02 g/liter	0.04 g/liter	1.2
47.0 g/liter	21	G	0.2 g/liter	0.4 g/liter	0.8
200.0 g/liter	16	G	0.0 g/liter	2.0 g/liter	1.0
0.17 g/g	18	G	0.000 g/g	0 001 g/g	0.6

*Data obtained over a six month period.

†F, fluorimetric analysis; G, pulse height analysis.

‡Variation % $= \frac{\text{S.D.} \times 100}{\text{A.S.V.}}$

Table 9—COMPARISON OF MEASUREMENT CONTROL REPORTS FOR DECEMBER 1957 AND FEBRUARY 1959

Average standard value	December 1957		February 1959	
	Method*	Variation,† %	Method*	Variation, %†
0.14–2.4 ppm	F	36.0	F	19.0
0.11–1.8 ppm	F	27.0	F	33.0
34.0 ppm			G	6.4
0.55–0.62 g/liter	C	1.6	G	1.3
1.6 g/liter	C	2.6		
1.1–3.3 g/liter	C	1.6	G	1.2
39–47 g/liter	T	0.36	G	0.9
390–200 g/liter	T	0.02	G	1.0
0.20–0.17 g/g	T	0.36	G	0.6

*F, fluorimetric analyses; G, pulse height analysis.

†Variation % $= \frac{\text{S.D.} \times 100}{\text{A.S.V.}}$

Table 10—EFFECT OF MEDIUM ON COUNT RATE

Medium*	Count, 6 min	Available count, %
H_2O	1226×10^3	100
5 N HNO_3	1226×10^3	100
12 N HNO_3	1216×10^3	99
82% by weight SiO_2	1182×10^3	96
75% by weight $Al(NO_3)_3 \cdot 9H_2O$	1113×10^3	90

*Equal amounts of enriched uranium as $UO_2(NO_3)_2$ present in all five samples.

fected significantly. The SiO_2 simulates the effects of filter muds which are mainly silicious ash. The aluminum nitrate was added as a solid and the main portion did not dissolve. Samples of these types would have to be spiked to allow for the effect of these interferences. Table 11 shows the count rate vs the form of enriched uranium.

SUMMARY

A procedure has been devised to determine the enriched uranium content of material obtained during salvage operations. Tables 1 through 9 show that on both production and control samples the results obtained by pulse height analysis have about 1.0 per cent variation. This is sufficient accuracy to replace the former chemical methods with a considerable savings in time and effort.

Table 11—COUNT VERSUS FORM OF ENRICHED URANIUM

Form*	Count, 6 min	Available count, %
$UO_2(NO_3)_2$	3309×10^3	100
UO_3	2695×10^3	81
UF_4	2544×10^3	77
U_3O_8	2367×10^3	72
U–Metal	1578×10^3	48

* Equal amounts of enriched uranium in each form.

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SS MATERIAL INPUT MEASUREMENTS TO THE ORNL POWER REACTOR FUEL REPROCESSING PILOT PLANT

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ABSTRACT

An interim pilot plant facility for processing power reactor fuels has been put into operation at Oak Ridge National Laboratory. This facility will investigate the modifications and additions required in existing solvent extraction technology to assure its usefulness in reprocessing the newer type civilian power reactor fuels and to undertake the development of new processes which may prove more economical and simple than the existing technology. The Purex Process is used for the solvent extraction process with various types of head-end treatments to prepare the irradiated fuels to a form suitable for solvent extraction.

S. S. Material input to the pilot plant is determined after the dissolved metal solution is transferred to the feed input measurement vessel. A study is currently being carried out to determine the over-all limits of error for measurements of the uranium and plutonium input. Vessel instrumentation and calibration are discussed and the results obtained from solution mixing, sampling and analytical tests for the feed input measurement vessel are presented.

1.0 INTRODUCTION

The ORNL Power Reactor Fuel Reprocessing Pilot Plant is engaged in a study to determine the over-all limits of error (95% confidence level) for the measurements of uranium and plutonium fed into the pilot plant. At the present time this study is concerned only with the feed input measurement vessel. Later we expect to extend our study to product withdrawal.

We have not processed any power reactor fuels as yet. At the present time we are trying to develop specifications for S. S. accountability measurement.

The pilot plant is comprised of two facilities which are connected by underground piping for inter-transfer of solutions (Fig. 1). Reactor fuels can be dissolved in the 3505 Building and pumped to the dissolver vessel in the 3019 Building. Metal can also be dissolved in the 3019 Building. From the 3019 dissolver, the dissolved metal solution is transferred to the feed input measurement vessel (S-2). After the volume of solution in this vessel is determined and the solution analyzed, a measured quantity of solution is jetted to a feed tank. The feed solution is processed through one cycle of solvent extraction in the 3019 Building, transferred to the 3505 Building and processed through further solvent extraction cycles. Plutonium and uranium products are withdrawn at Building 3505.

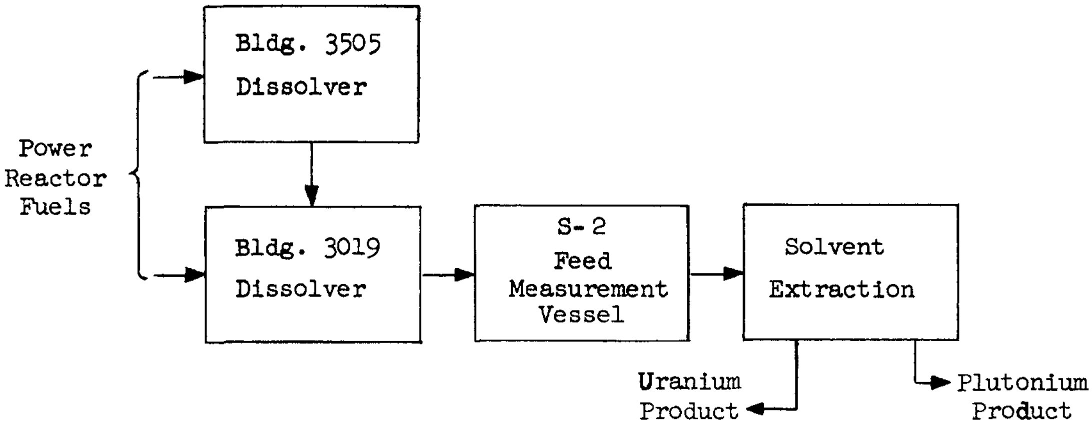

Fig. 1—Schematic representation of the ORNL Power Reactor Fuel Reprocessing Pilot Plant.

Concerning feed measurement, the following will be discussed:

1. Vessel instrumentation
2. Vessel calibration
3. Solution mixing
4. Sampling
5. Analyses

2.0 VESSEL INSTRUMENTATION

The instrumentation for measuring and recording liquid level and specific gravity of feed solution in S-2 is shown in Fig. 2. This is a 550-gal vessel with a reverse dish bottom and a straight dish top. The capacity of the vessel is about 1600 liters to the top of the straight-wall section.

Four air-purge probes extend into the vessel, two for specific gravity and two for liquid level. The specific gravity probes are separated 10.2 inches at the bottom of the vessel, while one liquid level probe extends to the bottom with the low pressure probe just inside the tank top. Purge air is metered through air rotameters at 0.2 scfh and introduced into the probes just above the tank top.

Pressure differential across each set of probes is sensed by dual transmitters, Foxboro Model 13A. Two panelboard-mounted Foxboro M-53 miniature recorders receive the output signals from the transmitters. Each recorder receives liquid level and specific gravity measurement.

A mercury-filled manometer mounted behind the panelboard can be plugged into the rear of each recorder to measure the output signal from each transmitter. This can be done to check the system in the event the recorder readings do not agree within one chart division. Two manometers mounted in the penthouse adjacent to the transmitters can be plugged into the transmitter input to take a direct measurement across either the liquid level or specific gravity probes. These manometers allow comparison of the pressure differential measured across the probes with the recorder values to determine which recorder is in error.

The vessel is also equipped with a thermocouple for temperature measurement.

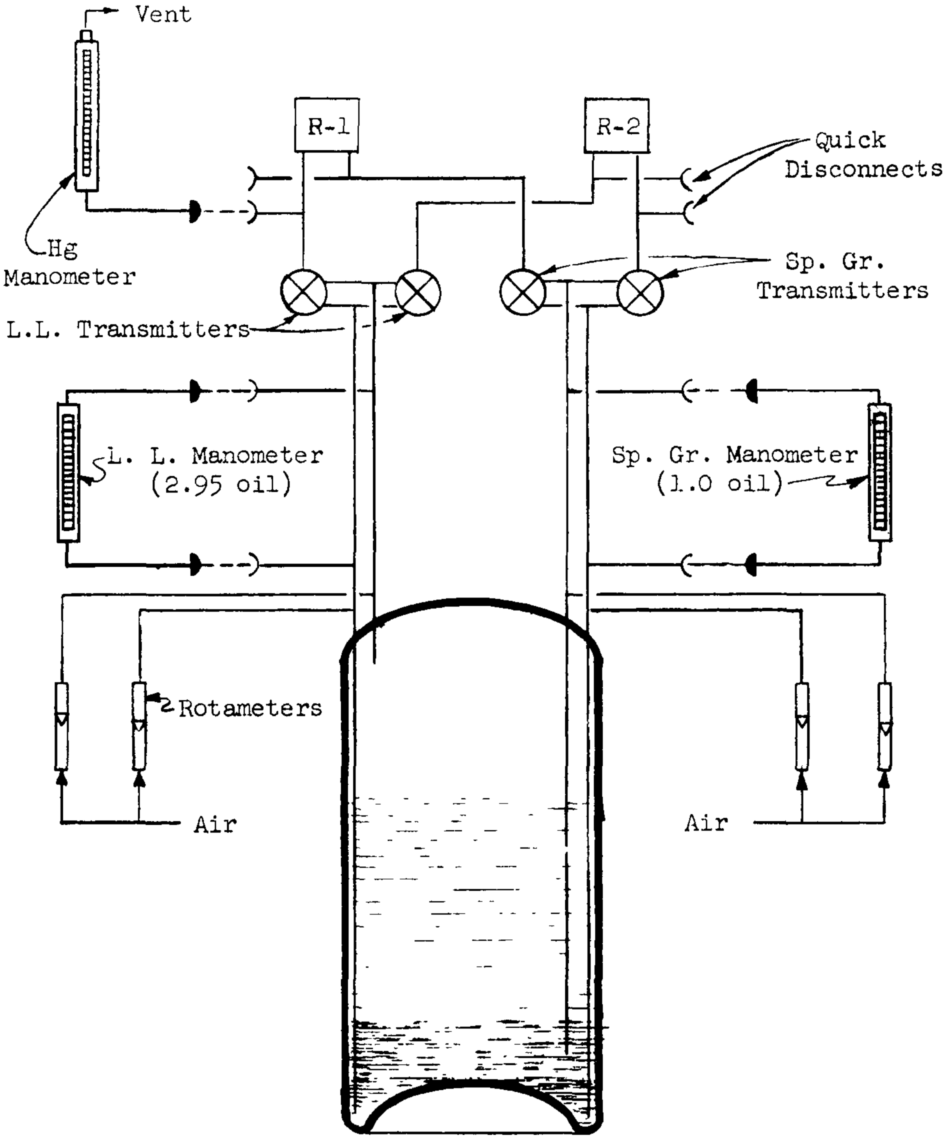

Fig. 2—Schematic view of S-2 instrumentation.

3.0 VESSEL CALIBRATION

The vessel was calibrated before the Power Reactor Fuel Reprocessing program was begun. This was done by adding weighed amounts of calibration fluid to the vessel and recording liquid level and specific gravity after each increment. After the vessel was filled, the calibration fluid was withdrawn from the vessel in weighed increments. Both water and non-radioactive uranyl nitrate solution of approximate feed concentration were used for calibration.

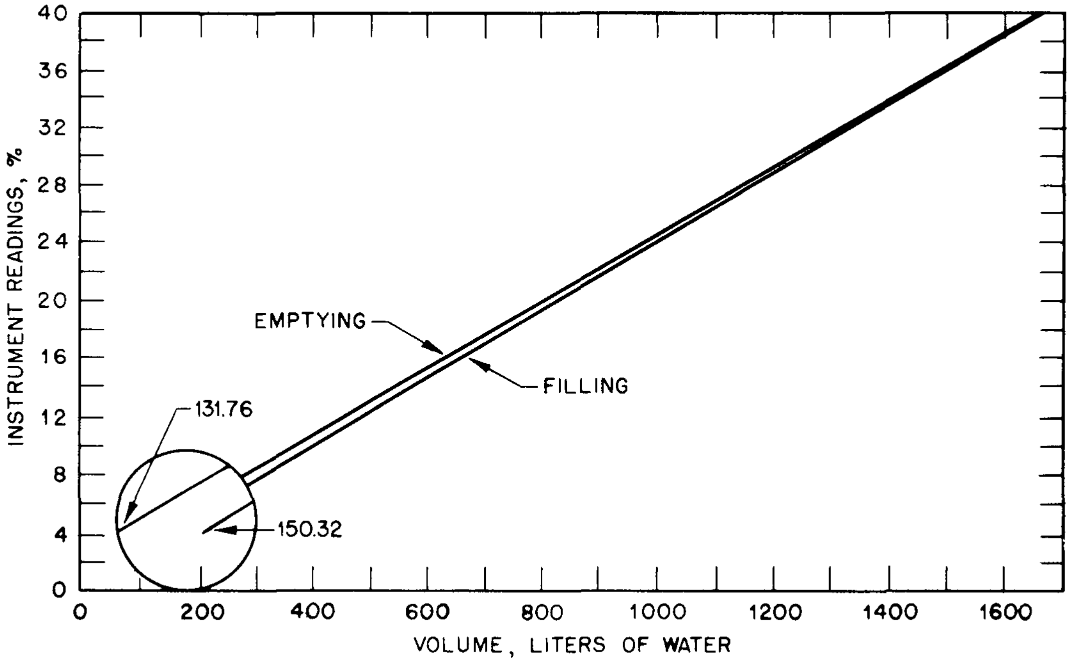

Fig. 3—S-2 calibration.

These calibration runs resulted in two calibration curves, one for filling the tank and one for emptying the tank (Fig. 3). There is an apparent hysteresis effect when emptying the tank. At the top of the curves, at a true liquid level of 40%, both curves give essentially the same volume. The filling curve gives 1663 liters while the emptying curve gives 1660 liters. However, at 4% liquid level, the difference in volume is 18.56 liters (150.32 for filling and 131.76 for emptying). When solution is transferred to S-2, the filling curve is used. When solution is removed from S-2, the emptying curve is used.

The coefficients for the curves were determined by a least squares fit. The curve for filling was computed from 46 calibration points, 20 being water calibration points and 26 being uranyl nitrate points. The curve for emptying was determined from 20 water and 25 uranyl nitrate points.

More scatter was obtained in the data taken when calibrating by emptying the vessel than when filling the vessel. The limits of error curves for both calibration curves (Fig. 4) show almost twice as much error for emptying the vessel.

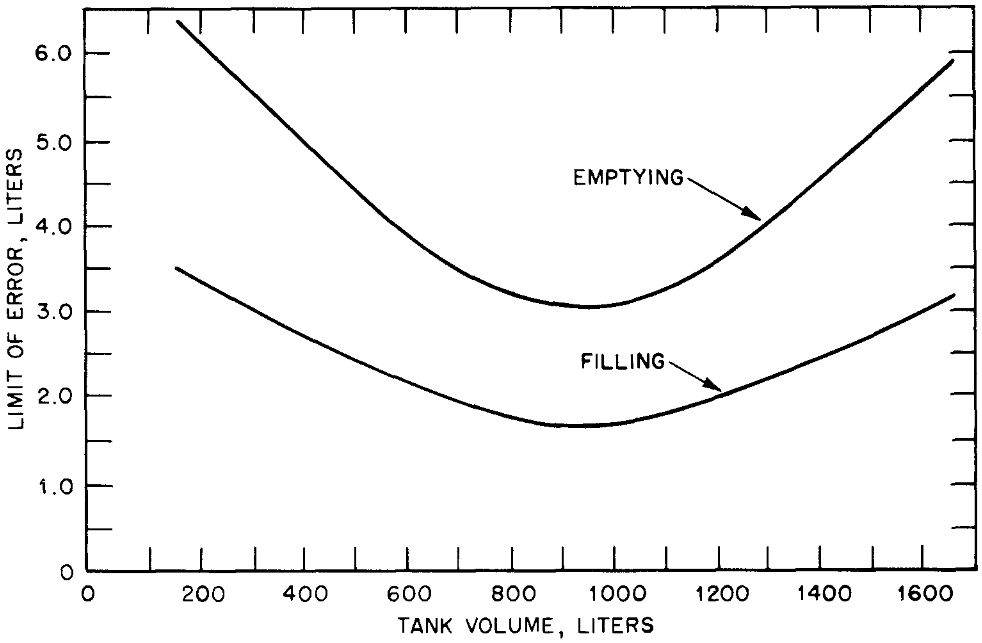

Fig. 4—Limits of error curves for S-2 vessel calibration.

The operating procedure for feed measurement is to fill S-2 to about 1400 liters volume. The solution is air sparged for two hours, sampled and the data are recorded. About 1000 liters of solution is jetted from the vessel, leaving a measureable heel of about 400 liters which is combined with the next feed batch.

4.0 SOLUTION MIXING

Solution in the vessel is mixed by air sparging. A series of tests were conducted to determine the required sparge time to assure solution homogeneity. Approximately 900 liters of uranyl nitrate solution containing 200 grams uranium per liter was added to the vessel. To this 250 liters of UNH containing about 360 grams uranium per liter was added followed by 240 liters of water.

The solution was sparged for 15-minute intervals and sampled after each sparge period. After 45 minutes of sparging, one-fourth of the solution was jetted from the vessel and the sparging periods were resumed. These tests were repeated until one-fourth of the original volume was sparged. By jetting one-fourth volumes it was felt that if stratification existed in the vessel, a new solution would be presented to the sampler and sparger at the tank bottom each time solution was jetted.

The specific gravity of the samples was measured in the analytical laboratory with a Westphal balance. The specific gravities are shown in Table 1. The change in specific gravities is insufficient to indicate nonhomogeneity. A change of 0.001 specific gravity unit is equal to about 0.75 grams per liter of uranium. Since our operating schedule permits, we arbitrarily sparge each batch of feed solution two hours before sampling.

During feed preparation samples were taken before and after transfer of solution from S-2 to continue to look for nonhomogeneity. This will be discussed later in Section 6.0.

Table 1

Specific Gravities of Solution Sparged vs. Sparge Time and Tank Volume

Starting Solution: 900 liters UNH Sp. Gr. 1.305
250 liters UNH Sp. Gr. 1.5
240 liters Water

Sparge Time, min	Relative Solution Volume			
	Full	3/4	1/2	1/4
15	1.258	1.258	1.258	1.259
30	-	1.258	1.258	1.260
45	1.258	1.259	1.258	1.259
60	-	-	-	1.259
75	-	-	-	1.258
90	-	-	-	1.259
105	-	-	-	1.259
120	-	-	-	1.259
135	-	-	-	1.259
150	-	-	-	1.259

5.0 SAMPLING

Solution is sampled from S-2 tank by airlifting the solution to a sample bottle outside the cell in a sample blister. The solution returns to the vessel. A test was made to determine the sampler recirculation time required to produce samples of uniform composition.

The sampler was recirculated for 5-minute intervals for a total time of 30 minutes. At the end of each period the bottle was removed and a second bottle was placed on the sampler tips long enough to obtain 10 ml of solution.

The samples were analyzed for uranium concentration by the ammonium thiocyanate colorimetric method. The range of the analyses was from 304 to 300 grams uranium per liter over the 30-minute recirculation period, indicating no significant effect of recirculation period vs. concentration. Since there is sufficient time during feed preparation, the sampler is recirculated 20 minutes for each set of samples taken. The recirculation sample bottle is discarded to eliminate the chance of submitting a sample containing either dilute or concentrated solution flushed from the sampler supply line.

It is emphasized that the range of 304 to 300 grams/liter of uranium was not obtained with run-of-the-mill samples, but rather with clear, clean nonradioactive solutions permitting bench-top analysis.

6.0 ANALYSES

In the first 2.5 months of this year 60 batches of feed solution were prepared to process four reactor fuel programs. The feed samples taken from S-2 tank were analyzed by the 3019 Building analytical control unit either by conventional bench-top methods when the samples emitted less than 1 roentgen/hr of penetrating radiation or by remote manipulators in the hot cells when the radiation level was greater than 1 roentgen/hr.

One dilution was made from an aliquot of each sample. Two determinations of uranium (ammonium thiocyanate colorimetric method) and two of plutonium (TTA extraction method) were made on each dilution. The two determinations were averaged and reported as an analysis on a single sample.

The feed solution sampling and analysis plans were changed as the program developed to yield information on four items important to analytical precision and bias. These plans were designed to give information on the following items for both uranium and plutonium analyses:

1. The sample variance.
2. The difference between analyses made by the control unit at two different times on the same sample, but not necessarily by the same analyst.
3. The difference between analyses made on the same sample by two different ORNL analytical units, one being the control unit.
4. The difference between analyses of samples taken before transfer and after transfer of feed solution from S-2 tank.

The following is a brief discussion of each item:

Initially four samples of feed solution were taken prior to transfer of the feed solution from S-2 tank to the process. To eliminate real "fliers" from consideration, it was required that the range of the four analyses be less than or equal to 7.25% of the average. If the analyses satisfied this requirement they were accepted; if not, four more samples were taken for analysis. Out of 28 sets of four samples taken from 21 batches of feed only one set was rejected for exceeding the range criterion without having a definite operational reason for rejecting the samples. Of the 21 sets accepted, the average range was 1.7% of the grand mean for uranium and 2.5% for plutonium. The pooled sample variance for uranium was 7.6 $grams^2/liter^2$, or 1.9 $grams^2/liter^2$ per analysis. For plutonium the pooled variance was 2.4 x 10^{-6} $grams^2/liter^2$, or 0.6 x 10^{-6} $grams^2/liter^2$ per analysis.

Six sets of four samples each were re-submitted to the control unit for re-analysis of uranium and plutonium. From these data it was evident that the precision was good; however, there was a difference between means for the first analysis and the second analysis. In other words the range for four samples was a satisfactory percentage of the average, but the averages were shifting up and down from the first to the second time that the samples were analyzed. This variability was much more significant than the sample variance.

Six sets of samples were submitted to other analytical units at ORNL to determine the bias between laboratories for uranium and plutonium analyses. At the present time there is insufficient information to determine if a significant difference exists. For uranium the average difference between laboratories was 2.7 ± 6 grams/liter and for plutonium was 0.0006 ± 0.0012 grams/liter.

Taking into consideration the need for information on the four items previously mentioned and analytical costs, radiation exposure to analytical personnel and sample load on the laboratory, a sampling and analytical plan was designed as follows:

1. Before transfer of feed solution two samples were taken. One sample was analyzed for uranium and plutonium and one was stored.

2. After transfer of feed solution two samples were taken. One sample was analyzed and one was stored.

3. If the range of both uranium and plutonium analyses was less than 5% of the average, the analyses were accepted and the stored samples were discarded. If the range was too great, the stored samples were analyzed and the first set of analyzed samples was discarded.

4. The two samples accepted were re-analyzed by the control unit and then stored for analysis by another laboratory (at time of writing these samples have not been transferred to another laboratory).

Twenty-eight batches of feed solution were prepared using this sampling and analysis plan. The results were not greatly different from those previously given. The pooled variance for sets of two samples was 11.9 for uranium compared to 7.6 for sets of four. The variance for the difference between the first and second time of uranium analysis for sets of two samples was 162 compared to 110 for sets of four.

It was disturbing, however, to notice that there was a definite difference in analyses of samples taken before transfer and after transfer. For uranium, the average difference was 8 ± 7.3 grams/liter and for plutonium was 0.004 ± 0.004 grams/liter. In fact, in checking with the operations group it was found that a slight leakage of either water or steam was diluting the feed solution. With such a slight leakage it is difficult for operations to determine whether or not the leakage has been stopped within the time available. Renewed efforts are being made to eliminate the source of dilution.

7.0 CONCLUSION

The over-all limits of error calculated for the first three programs are given in Table 2. These limits take into consideration precision and bias of volumetric and analytical measurements.

Table 2

PRFR Pilot Plant Feed Input Measurements

Program	U, kg	Pu, grams
1	5,386 ± 200 (3.7%)	2,028 ± 57 (2.8%)
2	---	1,120 ± 145 (12.9%)
3	1,496 ± 58 (3.9%)	1,518 ± 30 (2.0%)

Program 2 contained 5-10 times more plutonium per unit volume than the other programs and there was evidence that the first analyses may have been invalid. However, all analyses reported were combined, resulting in a much higher limit of error than would have been obtained had only the best values been used.

As the study continues and more information is obtained, we expect to make corrections in our data, we expect these limits to become larger and we expect to get more information on the true state of affairs---both good and bad.

ACKNOWLEDGEMENT

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SOME STATISTICAL ASPECTS OF B-PID'S AND ENDING INVENTORIES

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1. INTRODUCTION

The B-PID is the basis for many decisions which may entail large expenditures of time, effort and money. From a statistical point of view, the data are not always used in the most efficient manner and the results contained in this paper are intended to suggest some new ways of considering the accounting structure that exists and of estimating inventories and B-PID's.

The B-PID is defined as the algebraic difference between the physical inventory and the book inventory for a material balance area and is intended to detect such things as physical loss, diversion and gross errors of measurement or accounting. The book inventory is equal to the beginning monthly inventory plus the receipts and minus the removals for the month, so that under ideal conditions, the book inventory should equal the ending physical inventory. In actual practice at HAPO, the ending physical inventory for any month is used as the beginning inventory for the next month.

The situation is somewhat analogous to taking duplicate analytical samples, using both samples to control the measurement variation, but using only one of the samples to estimate the process average. In other words if the book inventory is an unbiased estimate of the true inventory all of the data are not being used in the most efficient manner to estimate the true inventory. The physical inventory is used as the estimate of the true inventory when it is possible to form an unbiased estimate using both the physical and book inventories, which has a smaller variance. A further development of this concept will be introduced by means of examples.

Let us consider the data which are available for one month. The physical inventory at the end of the past month is used as an estimate of the beginning inventory for the present month. Call this y_0. The difference between the receipts and removals estimates how much material has been added to or removed from the material balance area. Call this x_1. Then the quantity $y_0 + x_1$ is called the book inventory and should estimate I_1 the true amount of material in the material balance area at the end of the month. The quantity y_1 is the physical inventory and this is an independent estimate of the inventory at the end of the month. Under ideal conditions with no measurement error present the book inventory $y_0 + x_1$ should equal the physical inventory y_1 or

$$y_0 + x_1 = y_1$$

$$y_1 - (y_0 + x_1) = 0.$$

However, in the situations we are considering there is measurement error present so that some form of statistical treatment is necessary. To this end we define the B-PID to be equal to

$$\text{B-PID} = y_1 - (y_0 + x_1) = w$$

and note that it has these statistical properties. If no gross error has been made, or no loss or diversion is present the "average" B-PID (or the expected value of the B-PID) should be equal to 0. Since the quantities y_1, y_0, and x_1 are independent in a statistical sense the variance of the B-PID is equal to $2\sigma_y^2 + \sigma_x^2 =$ var. B-PID. Then the statistic

$$u = \frac{\text{B-PID}}{\sqrt{2\sigma_y^2 + \sigma_x^2}}$$

may be used to test the hypothesis that the B-PID = 0.

The book inventory $y_0 + x_1$ or the physical inventory y_1 may be used as the ending inventory for the month. The physical inventory has a smaller variance than the book inventory y_1, so that it seems reasonable to use y_1 as the estimate of the ending inventory. However, there is another possibility, any linear combination of the form

$$z = k\,(y_0 + x_1) + (1 - k)\,y_1$$

gives an unbiased estimator of the ending inventory and from this family of estimators it is possible to determine one which has minimum variance. Let

$\sigma_y^2 = B, \sigma_x^2 = A$, then it can be shown that

$$\frac{B}{A + 2B}(y_0 + x_1) + \frac{A + B}{A + 2B}y_1 = \hat{I}$$

is the estimator which has the minimum variance. If we let A = Bc $\hat{I}$ can be written as

$$\hat{I} = \frac{1}{c + 2}(y_0 + x_1) + \frac{c + 1}{c + 2}(y_1)$$

$$\hat{I} = \frac{1}{c + 2}(y_0 + x_1) + \frac{1}{1 + \frac{1}{c + 1}}(y_1) \;.$$

Further, it can be shown that the

$$\sigma^2_{\hat{I}} = \left(\frac{c + 1}{c + 2}\right) B = \left(\frac{1}{1 + \frac{1}{c + 1}}\right) B$$

$\sigma^2_{\hat{I}}$ is then $100\% \left(\frac{1}{c + 2}\right)$ smaller than $\sigma^2_{y_1}$. As a numerical example suppose that

$$y_0 = 50$$

$$x_1 = 25$$

$$y_1 = 78$$

$$\sigma^2_y = 2$$

$$\sigma^2_x = 1$$

Then $c = 1/2$, and

and
$$\hat{I} = \frac{2}{5}(50 + 25) + \frac{3}{5}(78) = 76.8$$

$$\sigma^2_{\hat{I}} = \frac{3}{5}(2) = 1.2 = (1 - \frac{2}{5})\, 2 .$$

Here the variance of $\hat{I}$ is 40% less than the variance of y_1.

Suppose that data are available for a 4 month period, where no measurement error is present.

Month	Beginning Inventory	Receipts-Removals	Physical Inventory
1	10	10	20
2	20	5	25
3	25	-10	15
4	15	5	20

The final inventory I_4 is equal to

$$10 + (10 + 5 - 10 + 5) = 20, \text{ or}$$

$$20 + (5 - 10 + 5) = 20, \text{ or}$$

$$25 + (-10 + 5) = 20, \text{ or}$$

$$15 + (5) = 20, \text{ or}$$

$$20 = 20.$$

In a system such as this, where measurement error is not present, there are many ways to calculate the inventory at the end of n months. However, if we superimpose a measurement error on the values, where the measurement errors are from a table of random normal deviates with $\sigma = 1$, $\mu = 0$, we obtain

Month	Beginning Inventory	Receipts-Removals	Physical Inventory
1	9.144	8.195	19.724
2	19.724	7.285	25.379
3	25.379	-10.602	16.468
4	16.468	5.229	12.382

and the five estimates of the final inventory I_4 are

$$q_1 = 9.144 + (8.195 + 7.285 - 10.602 + 5.229) = 19.251$$

$$q_2 = 19.724 + (7.285 - 10.602 + 5.229) = 21.636$$

$$q_3 = 25.379 + (-10.602 + 5.229) = 20.006$$

$$q_4 = 16.468 \quad 5.229 = 21.697$$

$$q_5 = 21\ 382 = 21.382.$$

Any combination of the q's

$$I_4 = k_1 q_0 + k_2 q_1 + k_3 q_2 + k_4 q_3 + k_5 q_4$$

in which

$$k_1 + k_2 + k_3 + k_4 + k_5 = 1$$

will give an unbiased estimate of the final inventory. If we wish to minimize the variance of such an estimate, it is not readily apparent how the values should be combined since the q_i values are not independent nor do they have the same variances. It can be shown that the values

$$k_1 = 1/55,\ k_2 = 2/55,\ k_3 = 5/55, k_4 = 13/55,\ k_5 = 34/55$$

will give $\hat{I}_4$ a minimum variance. Then

$$\hat{I}_4 = 1/55 \left\{ 19.251 + 2\,(21.636) + 5(20.006) + 13(21.697) + 34(21.382) \right\}$$

$$= 21.302$$

is the best such estimate in the previously defined sense. Further the variance of $\hat{I}_4$ is equal to

$$\frac{34}{55}\,(1^2)\ .$$

Since $\sigma^2_{y_4} = 1$, this represents a 38% reduction in the variance of the final inventory. The manner in which the k_i's are calculated makes an interesting subject in its own right. However, it would be too time consuming to show the development of the algebra involved. Suffice it to say that the last constant

$$K_n = \cfrac{1}{1 + \cfrac{1}{c + \cfrac{1}{1 + \cfrac{1}{c + \cfrac{1}{1 + \cdots \atop \cdots \cfrac{1}{c + 1}}}}}}$$

where n c's occur in the denominator of this continued fraction, and the limit of this fraction is equal to

$$t = \frac{-c + \sqrt{(c+2)^2 - 4}}{2} .$$

If we let $\ell_1 = (1 - t)^n$, $\ell_2 = t\,(1 - t)^{n-1}$, $\ell_3 = t\,(1 - t)^{n-2}$, ..., $\ell_n = t\,(1 - t)$, $\ell_{n+1} = t$, we have an approximate solution which in most cases is very good and for n moderately large converges very rapidly to the exact solution. For instance when

$$A = B$$

as in the case we have considered

$$t = \frac{-1 + \sqrt{5}}{2} = 0.618$$

and 34/55 = 0.618 correct to three decimal places. For practical purposes it is fortunate that the approximate procedure leads to a solution which is easily carried out. Using the same data again suppose we estimate $I_1 = 20$, by

$$\hat{I}_1 = (1 - t)\,(y_0 + x_1) + t\,y_1$$

$$= (.382)\,(9.144 + 8.195) + .618(19.724)$$

$$= 18.813.$$

Then

$$\hat{I}_2 = (1 - t)\ (\hat{I}_1 + x_2) + t\ y_2$$

$$= (.382)\ (18.813 + 7.285) + .618(25.379)$$

$$= 25.654$$

estimates $I_2 = 25$.

$$\hat{I}_3 = .382(25.654 + (-10.602)) + .618(16.468)$$

$$= 15.927$$

$$\hat{I}_4 = .382(15.927 + 5.229) + .618(21.382)$$

$= 21.296$ so that this iterative approximate solution is in substantial agreement with the exact solution. Further the values 18.813, 25.654, 15.927 and 21.296 give estimates of I_1, I_2, I_3 and I_4 which are approximations to the best estimates possible for the ending inventories at the end of the first, second, third, and fourth months. In summary then the practical method for making the maximum use of the data in the sense that we have defined it is to determine

$$t = \frac{-c + \sqrt{(c+2)^2 - 4}}{2}$$

where $c = \dfrac{\sigma_x^2}{\sigma_y^2}$ and determine $\hat{I}_1$ by

$$\hat{I}_1 = (1 - t)\ (y_0 + x_1) + t\ y_1,$$

$\hat{I}_2$ by

$$\hat{I}_2 = (1 - t)\ (\hat{I}_1 + x_2) + t\ y_2$$

and so on. It is then obvious that this procedure would be easy to carry out in practice.

At this stage we haven't indicated how our ending inventory estimate enters into the B-PID structure. A natural way to define the B-PID for the n^{th} month is by

$$y_n - (\hat{I}_{n-1} + x_n) = z_n$$

where $I_{n-1} + x_n$ is the book inventory for the n^{th} month. There are other definitions which suggest themselves but in general they are scalar modifications which use the data in essentially the same way. Then we use the statistic

$$\mu_n = \frac{z_n}{\sqrt{\sigma^2_{z_n}}}$$

and accept the hypothesis that $z_n = 0$ if $t_{1/2\alpha} < \mu_n < t_{1-1/2\alpha}$. In this instance we have a greater power of discrimination using this B-PID as compared to the usual definition using the physical inventory when a true discrepancy exists because the variance of our estimator z_n is smaller.

If the differences in the book and physical inventories are due to measurement error and no true discrepancies exist the covariance between successive B-PID's as ordinarily defined is equal to $-\sigma^2_y$. This accounts for the negative serial correlation often experienced in studying the manner in which B-PID's behave. It is an interesting and important point that the correlation between the z_i values for successive months tends to zero. (Markedly so for most values of $c_1 \; n \doteq 4$).

Thus, the definition of the B-PID as suggested by the ending inventory estimate induces the important statistical property of independence between B-PID's for successive months. As a matter of fact this suggests another approach to the problem. Suppose we consider the B-PID's as linear forms in the variables $x_1, x_2, \ldots,$

x_n, y_0, $y_1, \ldots, y_n$, then how do we determine these B-PID's such that the B-PID's give a meaningful measure of discrepancy and such that the successive z_i's are orthogonal (independent) or are tending toward orthogonality with increasing n. It can be shown that this approach leads to the same solution for estimating I_n.

The properties of the B-PID's based on these "best estimates" of the ending inventory are interesting from the point of view of a control tool. As indicated before they have more power to discriminate between real and random discrepancies for a one month period, or for an abrupt change.

Suppose that there is a constant loss of Δ units/month. Then the expected value of the B-PID as ordinarily defined is

$$E(\text{B-PID}) = y_n - (y_{n-1} + x_n) = -\Delta$$

so that the B-PID's will fluctuate around the line

$$\text{B-PID} = -\Delta$$

and this fluctuation will be in something less than a random fashion due to the covariance between successive terms. However, the expected value of z_n will be equal to

$$E(z_n) = -\Delta\left(\frac{1-(1-t)^n}{t}\right) = -\Delta\,(B(t,n)) \quad .$$

It can be shown that $B(t,n) > 1$ for all $0 < t < 1$ so that the discrepancy tends to be more pronounced as the number of months increase, and in the limit $E(z_n)$ reaches the value

$$E(z_n) = \frac{-\Delta}{t} \ .$$

The value z_n tends to deviate farther and farther from zero when there is a loss or diversion. This is a desirable property and adds greatly to its sensitivity as a control tool. Further the deviations will tend to be random around the curve

$$z_n = -\Delta \left(\frac{1 - (1-t)^n}{t} \right)$$

which for most values of t ($n \geq 4$) will be about

$$z_n = \frac{-\Delta}{t} \quad .$$

A bias in a measurement where no random error is present means a result that is consistently higher or lower than the true value. In a measurement with random error present a biased measurement is one that is high or low on the average. If there is a bias δ in the physical inventories the

$$E(y_n) = I_n + \delta$$

where I_n is the true ending inventory. This means that the average physical inventory estimate has an average value equal to the true inventory value plus an amount equal to the bias in the measurement. It can be shown that

$$E(\hat{I}_n) = E(y_n) = I_n + \delta$$

so that

$$E(w_n) = E(z_n) = 0$$

and there is little to choose between the ending inventory estimates in this regard. If there is a bias ϵ in the receipts minus removals value, this is algebraically equivalent to the case where a diversion of the amount ϵ per month is present so that

$$E(w_n) = -\epsilon$$

$$E(z_n) = -\left(\epsilon/t\right)(1 - (1-t)^n),$$

since $$B(t\,,n) = \frac{(1 - (1-t)^n)}{t} \geq 1$$

z_n will be more greatly affected. The purpose of B-PID's as a control tool is to discover discrepancies, and biases represent an assignable cause which should be corrected. In other words, under the hypothesis that there has been no loss, diversion, error or bias, $\hat{I}_n$ makes the best use of the data. When the hypothesis is not true z_n has a higher power to detect this.

There are several ways that the variance of the B-PID may be found. If we have good estimates of all the variances of all the measurements that go into the B-PID, we may compose the variance of the B-PID by taking these into account. Some recourse may be needed to statistical tools like the propogation of errors and such. If the values of σ_y^2 and σ_x^2 are then known or can be estimated then the variation in the B-PID's are easily found, since

$$\text{var } w_n = 2\,\sigma_y^2 + \sigma_x^2$$

$$\text{var } z_n = (1 + t)\,\sigma_y^2 + \sigma_x^2\,.$$

A technique which is often used in quality control work is to estimate the inherent variation by using 1/2 the mean square of successive differences or to use the standard deviation as estimated by the moving range divided by the d_2 factor for $n = 2$. These tools are especially useful when there are trends in the data due to factors like a steady loss state. It can be shown that

$$E(z_n - z_{n-1})^2 \sim 2\,\sigma_z^2$$

even when this steady loss state is present. However, this technique cannot be applied to estimating the inherent variation in the w_n values since

$$E(w_n - w_{n-1})^2 = 6\,\sigma_y^2 + 2\,\sigma_x^2$$

$$= 2\,\sigma_y^2 + 2\,\sigma_w^2 .$$

However, if we know the ratio $c = \sigma_x^2 / \sigma_y^2$ we can use this method to solve for σ_y^2 and σ_x^2 and compose the variance of $\sigma_{w_n}^2$ by

$$\sigma_{w_n}^2 = 2\,\sigma_y^2 + \sigma_x^2 .$$

In summary then, it can be said that the use of the best estimate of the beginning inventory based on all previous data, instead of the observed beginning inventory results in B-PID's having advantageous control properties. A theoretical investigation shows that these B-PID's have (1) a smaller inherent variance, (2) a greater power of discrimination between real and random discrepancies, (3) a greater sensitivity in the detection of trends or measurement biases, and (4) statistical independence between successive observed values of the B-PID in the absence of biases or trends.

The results have been empirically tested by creating models using error terms from a set of random normal numbers and observing how well the experimental and theoretical results agree. Further, the techniques have been applied to past historical data. In both cases there is excellent agreement between the way the techniques behave and the theoretical way they are supposed to behave.

DETERMINATION OF TOTAL AND ISOTOPIC URANIUM ON REACTOR ELEMENT SURFACES BY ISOTOPIC DILUTION, MASS SPECTROGRAPHIC ANALYSIS

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The fission recoil range of uranium in Zircaloy requires that U^{235} in the Zircaloy cladding of fuel elements be at a minimum. Since the early detection of a fuel rupture is based on the observation of fission products in the coolant stream, uranium from other sources would limit the sensitivity of these fuel defect detectors. For this and other reasons, a method was developed for the detection and isotopic assay of minute quantities of uranium in Zircaloy, steel, and other reactor materials.

Facilities

Due to the extremely low level of uranium in this analysis, special facilities were required for the preparation and processing of samples. Ideally, all equipment and the work area itself should have no past history involving uranium. Since this situation ordinarily is not practical, a description of the conversion of routine laboratories to this work is in order.

Two laboratories were selected for this use in which the history of past uranium processing was at a minimum. All equipment was removed from the rooms leaving only the laboratory furniture. After a thorough scrub down, these rooms were completely repainted. The floors were sealed with cocoon strip coat paint (1) and access is limited to only those persons performing the analysis. Personnel using the facilities are required to change into new footwear and laboratory coats stored in the clean area. All equipment then placed in service in this area is either new and/or sprayed with a clear lacquer to prevent the spread of loose contamination.

Samples submitted for analysis are screened prior to admission to the laboratory to prevent the possibility of cross contamination from unusually large amounts of uranium. This screening may consist of either alpha count analysis of U^{234} accompanying U^{235} or a fluorimetric analysis as described by Centanni, Ross and DeSesa (2) in which uranium is determined after a preliminary extraction separation by measuring the fluorescence of uranium in fluoride fusion under ultraviolet light. Any equipment used in processing samples of high U^{235} enrichment is removed from service to prevent cross contamination of subsequent samples. While these precautions may seem unusually severe, it must be remembered that this facility is intended for the processing of samples containing as little as 1×10^{-8} grams of uranium.

The observation of these precautions has permitted the successful operation of these laboratories in the immediate vicinity of other laboratories processing macro quantities of uranium. The total blank for the chemical separations method which includes reagent and enviornmental uranium is in the order of $.6 \times 10^{-9}$ grams of uranium.

Zircaloy specimens and Zircaloy clad fuels which have been corrosion tested in high temperature water acquire a tenacious (acid insoluble) surface oxide film. This surface film may be removed prior to analysis by a vacuum-annealing technique in which the surface oxide is caused to diffuse into the base metal thus exposing acid soluble metal. The vacuum furnace must be thoroughly cleaned and tested for uranium before use. The simple expedient of annealing previously analyzed Zircaloy coupons permits an evaluation of the degree of uranium transfer in the vacuum furnace. Zircaloy monitor coupons are routinely annealed with each batch of samples to note any environmental change in the vacuum furnace. Any transfer of material between areas is done with care to prevent contamination. New polyethylene sheeting is used to wrap all material and clean rubber gloves are used in all handling operations.

The vacuum annealing sequence is 4 hours at 650°C and $7x10^{-6}$ in. of mercury, then 6-1/2 hours at 730°C. Samples are then cooled in vacuum.

Special Reagents

In order to maintain low and consistent blank values, certain precautions must be observed in the preparation of reagent chemicals.

1. C.P. nitric acid is redistilled in an all glass still prior to use.
2. Distilled water of adequate purity has been prepared by either double distillation or mixed bed deionization (3).
3. The etch solution for zirconium and Zircaloy is prepared to be:

   5v/o C.P. hydrofluoric acid
   45v/o redistilled nitric acid
   50v/o redistilled water

   This solution is stored in clean polyethylene bottles.
4. Sample containers and other equipment. New polyethylene bottles are presoaked for 4 hours in 8M nitric acid and are then thoroughly rinsed with redistilled water before use. These bottles are not reused. This practice of presoaking and single use is observed for all other equipment when at all practical. Transfer pipets, polyethylene liners and similar equipment is used only once, while platinum ware and other equipment of this nature is most carefully cleaned.

Sample Preparation

A. Nitric Acid Wash (surface uranium)

Prior to sampling, the fuel element specimen is washed in 50% nitric acid to remove any loose surface uranium. The measured volume of wash solution is kept to a minimum to avoid unnecessary evaporation in the subsequent analysis. After a 5 min. agitation of the fuel element in the wash solution at room temperature, the nitric acid wash solution is transferred to a clean polyethylene sample bottle by use of a glass transfer pipette.

B. Surface Etch (subsurface uranium)

Analyses of subsurface uranium (~0.1 mil depth) have been performed by a controlled etch of the fuel cladding. The precorrosion tested fuel element or the vacuum annealed fuel element is immersed in a measured volume of the acid etch solution for a measured time interval. At the end of the etch period, the fuel element is immediately quenched in distilled water. The depth of cladding removal is then calculated from a zirconium analysis of the etch solution and the measured surface area of the fuel element. For the etch solution given, a 30 sec. exposure of 100 cm^2 surface/90 ml acid volume will remove 0.1 mil with a standard relative error of ± 20% at the 95% confidence limit. Deeper penetrations will not proceed at this same rate due to solution heating which increases the rate and acid depletion which decreases the rate.

These volume to surface area relations may be maintained in various ways depending upon the physical size and shape of the fuel element. Long slender elements have been etched in appropriate lengths of new polyethylene tubing closed at one end by means of a jumbo screw clamp. At the end of the etching period, the solution is then drained into a clean sample bottle. Plate type fuel elements have been etched in steel trays lined with new polyethylene sheet. The etch solution may then be sampled in a clean polyethylene wash bottle altered so as to draw in rather then discharge solution.

C. Localized Samples (particle inclusions)

Small selected areas have been sampled after metallographic examination disclosed abnormalities. The corrosion tested surface was broken by a prick punch and the selected area was encircled with a wax pencil. Successive one drop additions of the etch solution were made to the selected area and then transferred by means of a plastic pipet to a sample bottle and made to volume. Areas as small as 2-3 mm in diameter were sampled and analyzed for isotopic enrichment.

CHEMICAL SEPARATION FOR MASS SPECTROMETER ANALYSIS

In addition to the special reagent chemicals mentioned previously, the separations procedure requires:

1. Redistilled diethyl ether
2. Ammonium nitrate solution - prepared by diluting the redistilled nitric acid to 10M and bubbling ammonia gas through the solution until a basic test is obtained with pH paper. The excess ammonia is then removed by boiling the solution. The final solution is then made 2M with redistilled nitric acid.

All glassware such as centrifuge cones, stirrers, pipettes, and reagent bottles are presoaked in 50% nitric before use. Pipettes and cones are discarded after one use to avoid cross contamination of samples.

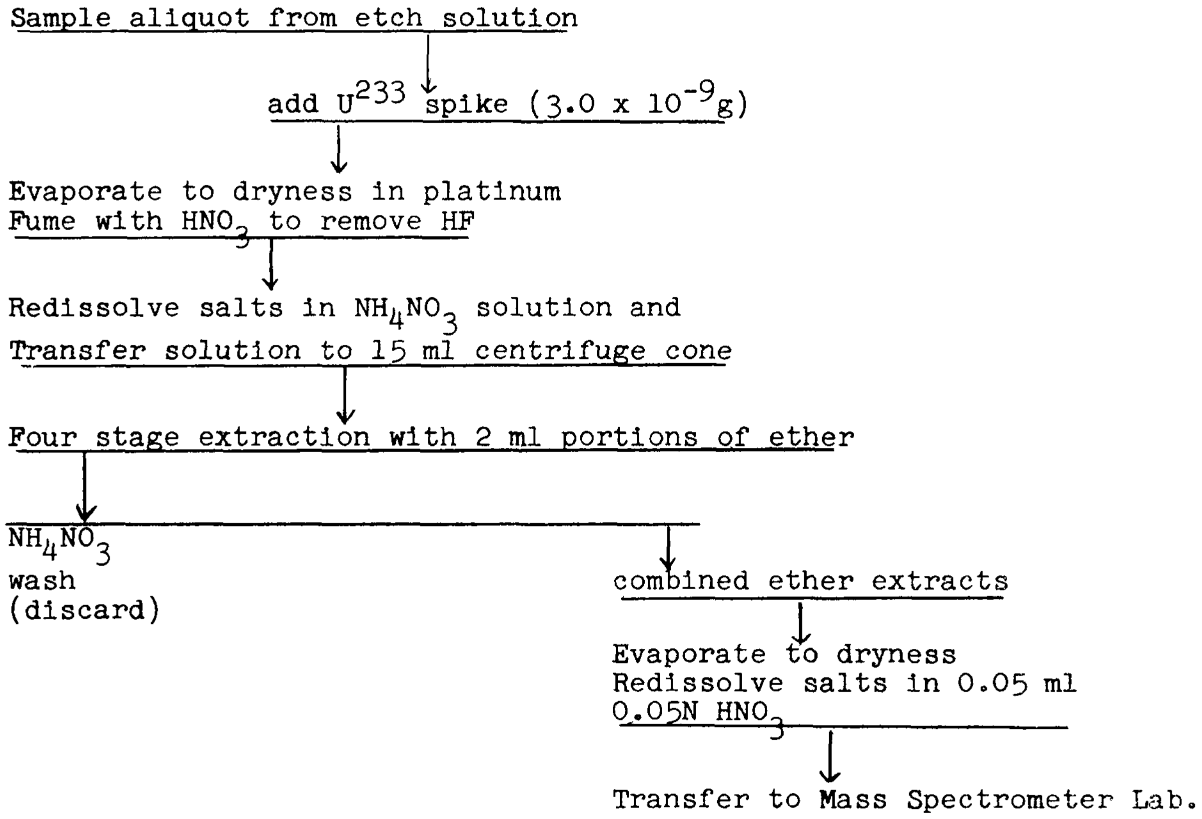
Procedure

A simplified diagram of the chemical separation procedure is shown in Figure I. This separation is explained in more detail in Reference (4).

The final solution from the chemical separation is transferred to the Mass Spectrometer Laboratory where the isotopic assay is performed. The isotopic dilution method is a powerful technique in analysis of microgram quantities of material. A quantitative extraction of the total uranium, after the U^{233} spike has been added, is not necessary. The reason is that all the uranium isotopes are recovered in proportion to the U^{233} spike. Assuming no large amounts of contamination by uranium during chemical separations, the final ratio of U^{233} to the other uranium isotopes, along with the accurately known amount of U^{233} spike, permits the estimation of absolute amounts of U^{235}, U^{238}, etc. Details of the mass spectrometer analysis have been reported by White, Collins and Rourke (5,6).

Figure I

Diagram of Chemical Separation Procedure



Accuracy

There are no accepted standards for uranium in zirconium or Zircaloy at these concentrations. However, comparison data has been obtained for the isotopic dilution-mass spectrometer method vs the fluorimetric method. This comparison of the two methods in Table I shows an agreement with a standard relative error of ± 20% at 95% confidence limits.

Table I

Fluorimetric Analysis
vs
Isotopic Dilution-Mass Spectrometer Analysis for Uranium in Zircaloy

Sample	Total U (ppm) Fluorimetric	Total U (ppm) Mass Spec	% U^{235}
1-3	2.15	2.37	0.77
1-10	1.67	1.64	0.77
1-17	3.93	2.88	45.7
2-5	1.17	0.67	1.10
2-25	227.6	203.0	93.0
2-26	2213.0	1752.0	93.0
3-4	1.29	0.63	5.03
3-7	0.79	0.60	1.89
3-10	48.6	56.2	-
4-4	1.77	1.23	1.16
4-14	4.50	3.96	79.0
4-15	18.9	15.5	87.8
5-4	1.27	1.23	0.86
5-13	1.74	1.34	9.6

Precision

The precision of the isotopic dilution method is illustrated in Table II.

Table II

Precision of Isotopic Dilution-Mass Spectrometer Method

Sample No.	Aliquot (ml)	Total U $g \times 10^{-9}/ml$	U^{235} %
1	1	8.73	45.25
	2	8.45	45.50
	5	8.32	45.25
2	1	10.5	2.94
	2	10.4	2.92
	5	10.4	2.96
3 (blank)	1	0.59	1.03
	2	0.66	1.95
	5	0.66	0.85

This data in Table II indicates that 1×10^{-8} g of uranium can be determined with a precision of ± 2 relative per cent.

Acknowledgment

The analytical method reported in this paper involves the skills of several groups at KAPL. I would like to give credit especially to Leo Collins and Frank Rourke of the Mass Spectrometer Laboratory and to Joseph Rynasiewicz, Vivian Consalvo and Merle Jones of the Chemical Laboratory.

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REDOX SS MATERIALS BALANCE TEST

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INTRODUCTION

There has long existed a need for improved measurements and controls of nuclear materials in a chemical separations plant. Since there have been few irradiating reactors and separations plants, the number of people interested in this problem is small. With the advent of programs for the processing of spent fuel from power reactors, there has developed a need to better define the reliability of nuclear materials measurements and controls in a chemical separations plant. The type of information needed may be divided into three categories:

1. Total measurement reliability, i.e., sampling, volume, and analytical measurements.

2. Plant material balance data for discrete quantities.

3. Comparison of plutonium values based on reactor calculations with measured values obtained after dissolution.

To provide this type of information a large scale SS Materials balance test with an extensive measurement program was conducted at a Hanford separations plant. This paper describes the design, results, and conclusions of this test.

The test was designed to minimize all recognized variables either inherent in the process or in the measurement and control system. In order to obtain the highest precision and accuracy practical, and also to establish experimental evidence of the validity of plant measurements, an extensive analytical, sampling, and volume measurement program was carried out. The methods for either eliminating or minimizing process and measurement variables that would have a direct effect on the results of the test were as follows:

1. Process

   Inventory accuracy and undetected wastes were considered as the two potential sources of processing error that would significantly affect the material balancing of the test. The effects of inventory error were essentially eliminated by processing the test material as an entity; that is, the beginning and ending inventory were made essentially zero. Therefore, the large estimating uncertainties associated with partially dissolved fuel elements and decontamination column inventories were eliminated. Each process vessel was flushed in a like manner both before and after the test. SS Materials that had accumulated in these flushes were measured by the most accurate techniques practical.

Possibilities for undetected waste losses were minimized by an absolute measurement control of all exit streams. For continuous large volume streams, such as cooling water, samples were taken continuously or at very frequent intervals.

2. Sampling Programs

Sampling errors can be generated from three sources: agitation of the solution, sample line circulation, and actual handling of the sample. The sampling program was designed not only for the elimination of these variables but, in addition, for studying their time dependency relationships. Logical questions for the sampling of a concentrated metallic ion solution from a vessel approximately twenty feet below the sampling port are; first, how long do we agitate the solution for homogeneity and, second, how long do we circulate the solution in the sample line before a representative portion of the solution is available in the sample port. It must be remembered that a single sample may represent as little as 1 part in 20,000,000. To provide a guarantee of sampling validity for the test, and as a means for studying the sampling characteristics, the sampling plan shown in Table 1 was carried out.

Table 1

Tank	No. of Samples per Batch	Remarks
H-7 Dissolver Solution	3	Three one-milliliter samples taken with eight minutes of circulation between samples.
Waste	2	Two two-milliliter samples with fifteen minutes of circulation between samples.
L-6 Pu Product Solution	6	One sample of L-6 tank prior to loadout of each Product Can. One sample of L-6 tank per Product Can loadout with fifteen minutes of circulation prior to each sample.
Product Cans	1	One thief sample of each Product Can.
E-12 U Product Solution	2	Two samples taken with fifteen minutes of circulation between each sample.

In addition, large samples were obtained of all other waste streams, such as process condensate and cooling water, leaving the building.

3. Analytical Program

As with any analytical laboratory, the possibilities for occurrence of random and fixed errors are many. There are variables in the analyst, reagents, instruments, equipment, techniques, and even in accepted conversion factors. Potential errors, both random and fixed, were either controlled or eliminated by the following analytical program:

> Random errors were controlled by duplicate and independent assays of each sample by different technicians. Multiple samples of each batch were of considerable help in the reduction of random errors. Except for several special analyses for comparative purposes, routine Redox plant analytical methods, equipment, and technicians were utilized throughout the test.
>
> Fixed errors were eliminated by increasing the coverage of the Hanford analytical standards program. In this program, known standards of simulated process samples are submitted to the laboratories for analyses, utilizing the identical procedures to analyze normal process samples. The analytical results of the process samples are then corrected by the differences between the recovered and known assay values of the standard.

The necessary assays for each material balance sampling point are provided in Table 2.

Plutonium Alpha Activity by Radio Chemistry

Total alpha activity of the dissolver solution is determined by mounting a micro-measured aliquot of sample directly on a stainless steel disc and counting the number of alpha disintegration for a given time interval in an Alpha Simpson Proportional Counter. A net plutonium alpha count is obtained by making minor corrections for uranium (~1.5%) and americium (~2%).

Americium is also determined by its alpha activity after a series of extractions and purifications. The uranium alpha contribution is determined from its specific activity constant.

Uranium Analysis

Dissolver Solutions

Two independent methods were used for the determination of uranium concentration:

(1) Extraction - X-ray Photometer Method

The uranium from a micro sample of dissolver solution is quantitatively extracted into an organic solution. The organic solution is then compared to a standard for the absorption of an X-ray beam.

(2) Density Method

This is the normal processing and material balance uranium measurement. A density is determined from the specific gravity of the solution measured by the falling drop viscosity method and corrected by the nitric acid content, which is measured by pH.

For specific batches, method 1 appears more reliable because of its selectiveness in the extraction step. The latter is highly dependent on acid concentration. Comparative material balances indicated however, only minor differences.

Plutonium and Uranium Product Solutions by X-ray Absorption

Both plutonium and uranium product solutions were measured by their X-ray absorption as compared to standards.

Table 2

LABORATORY ANALYSIS

Sample	Constituent	Method	Remarks
H-7 Dissolver Solution	Total Alpha	Radio-assay	Two dilutions -4 discs by each analyst on each of the three special samples.
	Am-Cm	Radio-assay	One dilution, two discs per analyst on each sample.
	HNO_3	pH-Meter	Single analysis on each sample.
	SpG	Viscosity (Falling drop)	Single analysis on each sample.
	U	Calculated from density	Calculated U for each sample.
Waste	U Pu	Fluorimetric Radio-assay	Duplicate analysis on each sample.
L-6, Pu Product Solution	Pu	X-ray	Duplicate analysis on each sample.
	SpG	Viscosity Falling drop	Single analysis on each sample.
	Alpha	Radio-assay	Duplicate analysis on each sample.
	Alpha/Pu element	Calculated	Calculated from each alpha and X-ray analysis.
E-12, U Product Solution	U	X-ray	Duplicate analysis on each sample.
	SpG	Viscosity Falling drop	Duplicate analysis on each sample.
	Impurities	Spectrographic	Every 5th sample.

Specific Gravity by Falling Drop Viscosity

The specific gravity is determined by the relative drop time of a given amount of solution in a known medium. Specific gravity is used for the conversion of solution weight measured by a water standard to actual solution volume.

Fluorimetric Determination of Uranium in Waste Streams

Microgram quantities of uranium in waste streams are measured by a fluorophotometer, the fluorescence generated by exposing uranium fused with sodium fluoride to an ultraviolet light.

4. Volume Program

The vessel calibration and relay instrumentation constituted the recognized volume variables. Material balance measurement vessels were calibrated by the weight increment technique. That is, an accurately weighed increment of water is added to a vessel and a weight factor recording made; this process is repeated until the whole of the vessel is calibrated. Instrumentation must always be controled therefore, each instrument was calibrated before the test and compared to a standard several times during the test. At H-7, the dissolver solution measurement point, and E-12 uranium measurement point, a standard manometer was operated in parallel to the existing one.

In addition, at H-7 and E-12, each manometer reading was verified by another reading five minutes later.

To maintain weight accuracy of plutonium product cans, a check weight, a secondary NBS standard, was made in conjunction with each can weighing. The can weight was corrected by the scale vs. the standard weight.

5. Test Material

The selection of test material consisted of a typical Hanford reactor product discharge. Exposure history was estimated to be within ± 2.5%. Uncertainties associated with reactor transmutation calculation was assumed equal to or greater than exposure history. If one were to consider all possible ramifications of reactor parameters, a ± 5% uncertainty estimate would have to be attached to the calculated plutonium values. In fact, one purpose of this test was to discover the reliability of a discrete normalized quantity of irradiated Hanford material. An attempt was therefore made to select a randomly distributed discharge for the purpose of normalizing reactor irradiation variables.

The uranium weight was based on statistically determined bare fuel element weight, adjusted for impurities, canning losses, and burnup in the reactor. Uranium weight reliability by the techniques used at Hanford is something better than (100 ± 0.1%).

Material Balance Areas (Reference Figure 1)

Two material balance areas are formed at the Hanford Redox Chemical Separations plant. The first is the initial dissolution measurement of irradiated uranium in the metal solution Preparation Area, with reactors as the input and H-7 tank as the output. The coating waste stream is within this area. Both uranium and plutonium are measured at the H-7 tank, and the measurement of SS materials at this point, in reality, constitutes a shipper-receiver measurement variance; by application of the (Pu/U)(U) ratio this difference is observed at point of charging.

The second is the extraction area, bounded by H-7 and (for uranium) E-12 tank and (for plutonium) L-6 i.e., PR product cans. Wastes are also accounted for within this area.

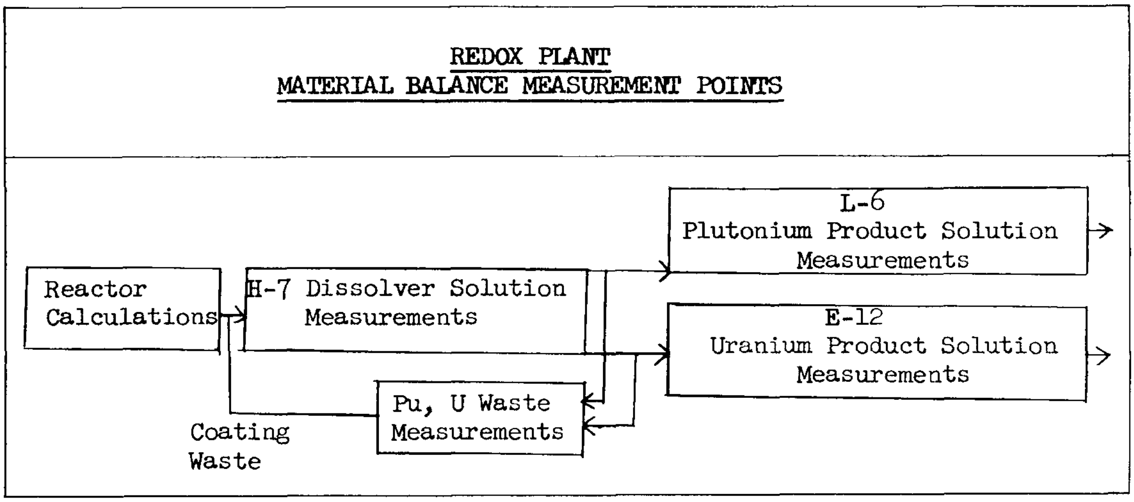

Figure 1

Uranium Material Balance

The uranium material balance obtained and amount of material involved are shown in Table 3.

Table 3

REDOX PLANT URANIUM BALANCE

Measurement Area	Analytical Bias Correction	Units Uranium
1. Reactor Area		100
2. a. H-7 Dissolver Solution by SpG	Corrected	99.16
	Uncorrected	99.94
b. X-ray Extraction	Corrected	98.40
	Uncorrected	98.50
3. E-12 loadout + waste	Corrected	100.57
	Uncorrected	100.47

Uranium Calculations

Uranium in both dissolver and product solution are calculated on the basis of uranium concentration multiplied by total aqueous volume. Impurities were subtracted.

The H-7 dissolver solution measurement of uranium, which is dependent on an aqueous volume measurement, demonstrates a recovery less than 100%. The quantitative plutonium measurement at this point by the aqueous volume method indicates this same tendency, concluding that volume measurement by remote weight factor instrumentation is presently biased low. This problem is being vigorously investigated by more frequent instrument calibrations, data analysis, and by research and development organizations.

Plutonium Material Balance

The plutonium material balance obtained and the amount of material involved are shown in Table 4.

Table 4

REDOX PLANT PLUTONIUM BALANCE

Measurement Area	Analytical Bias Correction	Units Plutonium
1. Reactor area		100.0
2. a. H-7 Dissolver Solution Volume Concentration	Corrected	103.2
	Uncorrected	103.8
b. Pu/U Ratio, U by SpG	Corrected	104.3
	Uncorrected	104.1
c. Pu/U Ratio, U by X-ray	Corrected	105.1
	Uncorrected	105.6
3. L-6 Pu Loadouts + Waste	Corrected	104.8
	Uncorrected	105.3

Quantitative Calculation of Plutonium

Receiving or Input Measurement at H-7 Tank:

Plutonium concentration is measured in the dissolver solution with respect to its alpha disintegration activity in relation to the aqueous median of the sample. Quantitatively plutonium can be calculated either, (1) as a function of the aqueous dissolver solution or (2) as a function of uranium charged to the plant by the reactors.

1. Plutonium alpha concentration multiplied by total volume.

2. Plutonium alpha concentration divided by uranium concentration, thus obtaining plutonium to uranium ratio. This ratio is then multiplied by the total uranium charged by the reactor. The reactor uranium values are actually the Hanford statistical onsite receiving weights, corrected for impurities, canning, and fission losses.

The quantitative plutonium alpha activity is then multiplied by the plutonium element loadout, divided by plutonium alpha activity loadout, thus converting the dissolver solution plutonium alpha activity to an elemental quantity.

Plutonium Product Solutions

Quantitatively, plutonium is determined from a net weight of each can and a specific gravity analysis. Plutonium is analytically determined by X-ray absorption as compared to a standard. For this test, each batch was sampled and, additionally, each transfer can (PR can) was sampled and analyzed. There were no significant differences established between the batch sample analysis and those of the transfer cans.

As shown by the data in Table 4, Redox plutonium values at all measurement points and by all three analytical techniques used are significantly higher than those predicted by the reactor calculations. On consideration of the uncertainties associated with reactor calculations and exposures, this discrepancy for the test of a relatively small batch of material appears to be within the expected order of uncertainty.

The most significant aspect of the plutonium balance is the degree of agreement between the initial H-7 dissolver solution measurements and the L-6 load-out values by the various methods. The ascertaining if such a balance is possible was one of the important criteria for the test. Results shown in tables 3 and 4 provide evidence that it is not only possible to obtain an accurate measurement of irradiated reactor fuels at the initial dissolver measurement point with all of the inherent measurement difficulties, sampling, heavy metal solution, high radioactivity, and volume, but it is also possible to confirm these quantities at the point of loadout.

Summary

1. Consistent, accurate, and representative sampling is possible at each of the measurement points. At each of the sampling points, it was observed that once a representative sample was obtained, additional samples were merely confirmation of the first sample after equilibrium. The two time-dependent sampling variables were determined highly related, but were repetitively consistent between batches. An analysis of variance of between and within analytical and sampling variations showed that between sample variation is negligible and that analysis of samples is the controlling factor.

2. Test evidence supported by previous examinations indicates that an analytical standards program is essential not only for the accuracy of analytical procedures, but also for the corrections of apparent drifts and trends. In each instance the application of standards data significantly improved the material balance.

3. Comparison of the first measurement whether analytical, sampling or volume, to the average of the multiple measurements, supports the obvious pure mathematical conclusion that multiple measurements increase precision but not necessarily accuracy.

4. Waste streams have the largest variance of all the accountability measurements made. However, this variance does not appear to be due to consistent errors and because of the small amount of material involved, the variances of the measurements are not a major problem for a material balance.

5. Individual plutonium-to-uranium ratios which are the quotient of two independent measurements appear to be as precise as the individual plutonium values. This lends support to the view that the ratio method is less subject to sampling variation than the volume-concentration method.

6. Accepting the Redox measurement as absolute, the test demonstrated the difficulties of either/or making accurate power measurements of transmutation calculations. The segregation of these two functions

in the Redox test was, of course, impossible; in actuality, they are undoubtedly confounded.

7. It appears entirely feasible to obtain an accurate initial measurement for material of unknown composition with later confirmation at the point of loadout. This will allow the chemical processing of power fuels in a large continuous separations plant confounded with other material with an accurate and assured basis for an imput measurement. As indicated before, the art of predicting reactor transmutation relationships has not advanced far enough to provide a base for economic or processing consideration. A firm, consistent, and reliable measurement method has now been evidenced by chemical separations plant. Process holdup, inventory problems, and other considerations in a separations plant are not limiting factors; what is needed is the complete dissolution of the material and movement to a measurements point. The plant balance as an entity will confirm and balance to the original dissolver solution measurement.

8. The Redox accountability test provided concrete evidence that within-plant material balances are possible for discrete quantities and that the initial input measurement of dissolver solution are representative of the material. Segregated processing of power fuels after the dissolution measurement will not be necessary. In addition, the close agreement between the Pu/U ratio method with L-6 loadout values supports the view that the use of input uranium values is superior to the accepted aqueous volume method for a plutonium production operation such as Hanford where integrity of the uranium values are known.

LIMITS OF ERROR IN WEIGHING

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INTRODUCTION

Techniques have been developed at M & C Nuclear, Inc., for increasing the accuracy of weighing and determining human performance in weighing. We have concluded that the frequency of human mistakes in weighing is not insignificant, that it should be measured and that it should be included in any estimate of and control of limits of error of weighing We base this on two extensive experiments we made last year

THE FIRST EXPERIMENT

To assure material to a corrosion resistance specification, a small coupon was taken from each fuel element and was subjected to an extended corrosion resistance test.

The customer required one half of this coupon for possible testing at his factory. Therefore the coupons, already small, were cut in half This resulted in coupons about $^5/_8$ in. square, $^3/_{32}$ in. thick, weighing about 3 g each.

Specifications were based on allowable weight gain per unit area. When they were applied to this size coupon the weight increase had to be held between limits differing by only 0.00070 g. We experienced an excessive number of rejections for this test. Eventually it was found that most of the rejections were due to weighing error. Some coupons even showed lost weight in the test.

We were using a pair of excellent Metler B-6 balances, reading to 0.00005 g The coupons were individually weighed before and after corrosion test, and readings were rounded off to the nearest 0.00010 g.

A number of coupons were weighed repeatedly on the two balances and it was found from the duplicate measurements that:

1. Each weighing observation had a standard deviation of about 0.00024 g. Therefore, weighing error alone could account for a 30 per cent rejection rate.
2. A difference between the two balances attributed to their starting and thereafter undisturbed zero settings of 0.00010 g.
3 About 5 per cent of the duplicate weighing observations showed serious discrepancies indicating operator mistakes.

When these three errors were taken from the data, the residual standard deviation was about 0.00008 g but this still was not accurate enough to guarantee meeting the specification

IMPROVING ACCURACY IN FIRST EXPERIMENT

The value of the rejected elements made it imperative for us to improve our weighing accuracy on the remainder of the fuel element coupons. The costs of weighing ceased to be an important factor. We could use any method of weighing that promised to be more accurate.

Since no better balances were available we decided upon a multiple weighing technique based upon a fully confounded 2^3 factorial experimental design.

DESIGN OF COUPON WEIGHING PROCEDURE

Seven coupons were weighed in eight weighings in the combination shown below:

	Weigh Coupons Numbered								Observed Weight
First Weighing			3		5	6		=	
Second Weighing	1					6	(7)	=	
Third Weighing		2			5		(7)	=	
Fourth Weighing	1	2	3					=	
Fifth Weighing			3	4			(7)	=	
Sixth Weighing	1			4	5			=	
Seventh Weighing		2		4		6		=	
Eighth Weighing	1	2	3	4	5	6	(7)	=	

For example, in the first weighing coupons numbered 3, 5, and 6 were weighed together, and in the eighth weighing all seven coupons were weighed together.

These particular combinations permit the eight weighings to be added algebraically to get the weight of any coupon independently of the rest. As an example, if we subtract the first four weighings from the sum of the last four we get 4 times the weight of coupon No. 4 by itself. The weight of coupon No. 4 appears in the last four weighings while the weight of each of the other coupons appears there twice, and also appears twice in the first four weighings which are subtracted. Thus the weight of all the other coupons are subtracted out.

Similiarly 4 times the weight of each of the other coupons can be obtained by adding the 4 weighings in which it appears and subtracting the 4 in which it does not appear. This procedure results in the following gains:

1. Because we can take the average of 4 weighing for each coupon, we can reduce random weighing error by 50 per cent.
2. We eliminate any zero bias of the balance by adding in such bias 4 times and subtracting it out the other 4 times.
3. In addition, we found that the configuration of the experiment made it possible to detect weighing mistakes. For example, if we find the sum of the seven computed weights do not equal the measured weight of the seven coupons the difference is an indication of error. (Also $\frac{1}{3}$ the sum of the first seven weighings should equal the last.)

RESULTS OF FIRST EXPERIMENT

When we checked the first 301 coupons weighed in this manner, we found of the 43 lots, 9 had differences of more than 0.00030 g and most of these were big differences indicating gross weighing mistakes.

Since each lot represented 8 weighings, our 21 per cent bad lots represented about 3 per cent weighing mistakes in the individual observations.

We cautioned the operators to be more careful but the next 43 lots showed 10 with weighing mistakes. No improvement whatsoever. Twenty-three lots more produced 5 with weighing mistakes, which was consistent with about 3 per cent operator mistakes per weighing observation despite presumably ideal weighing conditions in an isolated air-conditioned room.

These last two lots had been done with 8 weighings but only 6 coupons. The weight of the dummy seventh coupon was figured however, and its difference from zero gave a measure of the weighing error in the lot.

The data of the 15 lots with mistakes were analyzed to determine the probable magnitude of the mistakes. There was one mistake of a whole gram, despite the fact that readings were taken to five decimal places. However, 12 of the 15 mistakes occurred in the third decimal place.

The Metler B-6 balance has a different kind of scale beginning with the third decimal place and this apparently caused some human engineering difficulties. Only after cautioning the operators to particularly double check the third decimal place were we finally able to make a substantial reduction in operation mistakes.

FINAL RESULTS

The next lot of 45 showed 4 mistakes and the last lot of 46 showed 6 mistakes, a total of 10 mistakes in 728 weighings or $1\frac{1}{3}$ percent operator mistakes per weighing operation. Lots showing mistakes were reweighed. By utilization of this approach:

1. A considerable improvement in product assurance was achieved.
2. It was concluded the corrosion resisting process was in control. Coupons were made considerably larger on the next job.

THE SECOND WEIGHING EXPERIMENT

It was clear that even exercising the greatest of care in production weighing, when reading to six significant figures weighing mistakes still ran at the 1 per cent level. This fact disturbed not only Quality Control, but Accountability, and Manufacturing. An experiment was set up on a plant-wide basis to determine the limits of error, and corrective measures for production weighing. The plan was to measure:

1. Operator mistakes in production weighing under production conditions, and also with the operator knowing that he was being tested. This would measure the optimum influence of supervision.
2. The inaccuracies of the balances.

A set of secondary standard weights were prepared and weighed accurately by multiple weighings. (In an experiment similar to the first experiment.) These secondary standards were not in whole units of weight and were identified only by number.

An inspector went around three times a day to balances in use, and selected two production items which the operator had previously weighed. He recorded the weights which the operator had recorded and had the operator reweigh these two items with the proper secondary standards to bring the total weight to approximately 90 per cent of the capacity of the balance. Four such weighings were made. When it was possible (AB) included the two production items together; (A) and (B) included the production items separately, but (O) included standard weights only. A fifth weighing (OO) with standard weights only, at about 10 per cent of the capacity of the balance completed the check.

The use of the secondary standards whose weights were not known, obliterated for the operator the known results of the previously weighed production items. Since the total weight each time was brought to near the capacity of the balance, any zero and first order calibration error of the balance was cancelled out in analysis of the 2^2 factorial experiment represented by the first four weighings. The data were analyzed on our automatic digital computer which was programmed to subtract the secondary standard weights and then to calculate:

1. The average of each of the production weights;
   $a = \frac{1}{2}(AB + A - B - O)$
   $b = \frac{1}{2}(AB - A + B - O)$
2. The error;
   $e = \frac{1}{2}(AB - A - B + O)$
3. The zero bias of the balance;
   $z = O$
4. The first order calibration error of the balance, or the amount the balance is off when loaded near capacity to how much it is off near zero load;
   $c = OO - \frac{1}{4}(AB + A + B + O - 2a - 2b)$
5. The round off error;
   $r = 3$ in the place rounded off
6. The 95 per cent limits of error approximately;
   $S = 2(e^2 + z^2 + c^2 + r^2)^{\frac{1}{2}}$

RESULTS OF THE SECOND EXPERIMENT

We found that our balances were maintained in good calibration except when they were moved by the operator for relocation purposes. We then provided pedestal bases bolted to the floor to prevent this malpractice.

In general, the limits of error of the man balance combination showed a single weighing observation to have a standard deviation of 1.2 in the decimal place not rounded off. For example, if the balance could be read to two decimal places in grams and the results were reported rounded off to one decimal place, the standard deviation was about 0.12 g making the 99 per cent limits of error about 0.36 g.

Not much accuracy was gained by reading to the second decimal place, but quite a bit was lost if the first decimal was also rounded off.

As expected from our previous experimental results, the frequency of operator mistakes in production weighing ranged between 1 and 5 per cent. In weighing 60 fuel loads of about 300 g each by various operators, our check revealed 8 significant errors, 5 of about 0.6 g, 2 of about 1.2, and 1 of 14.9 g. One operator had as high as 20 per cent mistakes. He tended to read to the nearest 0.5 g. He promptly was brought into line with the rest by more supervisory attention, after the test. His gross mistakes were about the same as the others.

The number of mistakes in the check weighings themselves was about 1 per cent. This 1 per cent represents a target that perhaps could be achieved by careful supervision on single weighings where error detection is not built into the weighing procedure.

There was some evidence that the fewer the figures read on the balances, the lower the frequency of mistakes.

If you should wish to determine the frequency of your own operators mistakes, we recommend either a simple A, B, C, and ABC plan, or an AB, BC, CA, and Zero plan with or without secondary standards.

The least divisions of the balance should be read and rounded off one place.

These weighings may be summed up three ways to get double the weight of each A, B, C, free of zero bias, and the sums of the computed weights should agree with the combined weighings.

CONCLUSIONS

Under conditions of single weighings, without check, it seems unlikely that limits of error can be maintained with better than 95 per cent confidence.

Where many weighings are averaged together, mistakes of opposite value tend to cancel each other out. However mistakes do not appear to be distributed as in a normal distribution, and statistical methods of non-parametric nature should be employed to deal with them. Confidence statements cannot be made any better than the over-all frequency of operator mistakes allows.

Where weighing limits of error are required with 99 per cent confidence, all weighings should be double checked. A simple production method is to require a combined weighing to be made following a number of individual weighings. If the individual weighing values add up to the combined weighing value the individual weighings have been checked and can be used. It is desirable to keep a count or control chart of the frequency of mistakes caught by this system.

CONTROLLED-POTENTIAL COULOMETRIC ANALYSIS OF DISSOLVER SOLUTIONS OF VARIOUS REACTOR FUELS

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For the benefit of the persons in this group who are not analytical chemists it might be well first to describe briefly the controlled potential coulometric titration method. Then the results that have been obtained on the determination of uranium in synthetic dissolver solutions will be discussed.

When an analytical chemist is given a sample of material and asked to determine the amount of uranium present, he must have some information about the material for the intelligent selection of a proper method of analysis. Accuracy desired, approximate quantity of uranium, and elements present usually dictate the choice of the method. In the case of dissolver solutions the highly radioactive nature of the solutions also has a decided influence on the selection of a method. For reasons of high accuracy, selectivity, and ease of remote control the recently developed controlled-potential coulometric titration method has been selected for the analysis of dissolver solutions. This method dates back only to 1942 when Hickling[1] did the pioneering experiments. It was a laborious manual method until 1957 when Booman[2] developed an instrument that was a completely electronic potentiostat and integrator. This makes possible an automatic, in fact an almost push-button, analysis of various solutions The instrument that is used at ORNL is the same type but it has certain modifications including a transistorized power output.

The fundamental basis for this method is Faraday's law which states that when an electrode reaction takes place with 100 per cent current efficiency during an electrolysis, the amount of current consumed is a measure of the weight of the substance in the reaction The value of the Faraday has been determined very accurately by Craig and Hoffman[3] who found it to be 96,492 ± 3 coulombs. One Faraday of electricity is equivalent to 119 g of natural uranium. Enriched uranium has a lower atomic weight and therefore a lower equivalent weight which must be taken into account when analyzing enriched fuels. Therefore, when the total current, which at the start of the electrolysis may be 150 ma or more and decreases with time to 0.05 ma, is integrated it gives the equivalent of a substance either oxidized or reduced provided the reaction goes to completion.

Only two conditions are necessary for the accurate determination of uranium The first is to carry out the electrolysis so only one reaction takes place (i.e., the conversion of U(VI) to U(IV)) and secondly the total current for the electrolysis must be accurately integrated. The integrating device in the instrument is similar to the resistance-capacitance integrator as used in analog computers. The integration is accomplished by charging a large capacitor and reading its voltage on a readout device such as a potentiometer.

The instrument can be calibrated by means of a constant current which is measured precisely by the voltage drop across a standard resistor for a given length of time The integrated current by the instrument is measured on the readout device From the known coulombs of electricity fed into the instrument and the determined readout voltage, a coulometric factor is obtained for the readout voltage. Also precisely known quantities of uranium are titrated as a chemical standard Recently a study of the titration of 25 mg quantities of uranium was made. This is approximately the maximum amount of uranium this size cell can electrolyze practically. The aliquots of solutions were weighed and transferred to the electrolysis cell. The titration of ten aliquots showed a relative standard deviation of 0.05 per cent. The electrical standardization should agree with the chemical standards within 0.1 per cent

Figure 1 is a picture of the instrument showing the electrolysis cell and the digital voltmeter which is the readout device for the integrated current. The current meter is in the center of the instrument and can be set to stop the titration automatically at the background current of 50 μa. The applied voltage is adjusted by this potentiometer The electrolyte of $1\underline{N}$ H_2SO_4 is pretitrated to a background current of 50 μa and then the sample is added and deaerated with nitrogen. Figure 2 shows a diagrammatic sketch of the electrolysis cell. During the electrolysis the cell is purged with nitrogen to prevent any air-oxidation. The mercury pool is the cathode and the anode is a separated compartment of $1\underline{N}$ H_2SO_4. The reference electrode is the Ag-AgCl electrode. The potential of the electrolysis is rigidly controlled by the potentiostat circuitry of the instrument. The rate of stirring is quite important and the surface of the mercury pool must be vigorously stirred by means of the glass flat disk located at the interface of the mercury and electrolyte.

The potential for the electrolysis is chosen by first running a polarogram. This is accomplished by using a polarograph, which is a device for automatically recording voltage-current electrolysis curves of a solution by use of a dropping mercury electrode. Figure 3 shows a typical polarogram of a solution containing copper and uranium It will be noted that optimum potential for electrolyzing copper from the solution into the mercury pool should be −0.025 volt Then the electrolysis could be set to −0.3 volt to reduce U(VI) to U(IV). If the current is integrated for each of the electrolyses at these potentials, a measure of the copper and uranium present in the solution would be obtained. The reactions are shown in the following equations:

$$\begin{array}{lll} (1) & Cu^{++} \rightarrow Cu(Hg) & \text{Reversible} \\ (2) & UO_2^{++} \rightarrow UO_2^{+} & \text{Reversible} \\ & \qquad\qquad\downarrow & \qquad\downarrow \\ & \qquad UO_2^{++} + U^{+4} & \text{Net change} \\ & & \text{irreversible} \end{array}$$

In some cases where the copper or uranium ratios are different from those shown, a good separation of the electrolysis potential is not obtained In these cases the electrolysis is first carried to completion at −0.3 volt where both copper and uranium are reduced and then the copper is oxidized out of the mercury pool at +0.175 volts and the current is integrated As the uranium reduction is irreversible this gives a good measure for copper alone and the uranium is determined by difference This is the method now in routine operation for the analysis of highly radioactive homogeneous reactor fuel where copper is added as a recombining catalyst for oxygen and deuterium formed by radiolytic decomposition of the heavy water In fact three metals namely Cu, U, and Ni can be determined in the same 0.5 ml aliquot containing 5 mg of uranium of the fuel by proper selection of the electrolysis potential. Figure 4 shows the steps necessary for this analysis.

Table 1 shows the results obtained by analyzing synthetic HRT fuel. The relative standard deviation is 0 2 per cent. The analysis of the highly-radioactive HRT fuel has been carried out in the High Radiation Level Analytical Facility at ORNL by placing the electrolysis cell in the cave and the instrument outside the shielded area. Master slave manipulators are used for the necessary filling of the cell and adjustment of the stirring device. The relative standard deviation for the quality control samples over a period of several months has been 1.0 per cent. Table 2 shows these results. This includes all errors such as pipetting the samples and it is believed that this precision can be improved by improving the pipetting and mechanical techniques. The precision of the analyses outside the cave is always better than in the cave, however, better than 1 per cent precision should be realized in the cave.

Some work has been done to determine the effect of radiation on the method. Several analyses of known uranium solutions containing additions of highly radioactive manganese have been made. These solutions measured 40 r/hr which was about $\frac{1}{5}$ as radioactive as the HRT

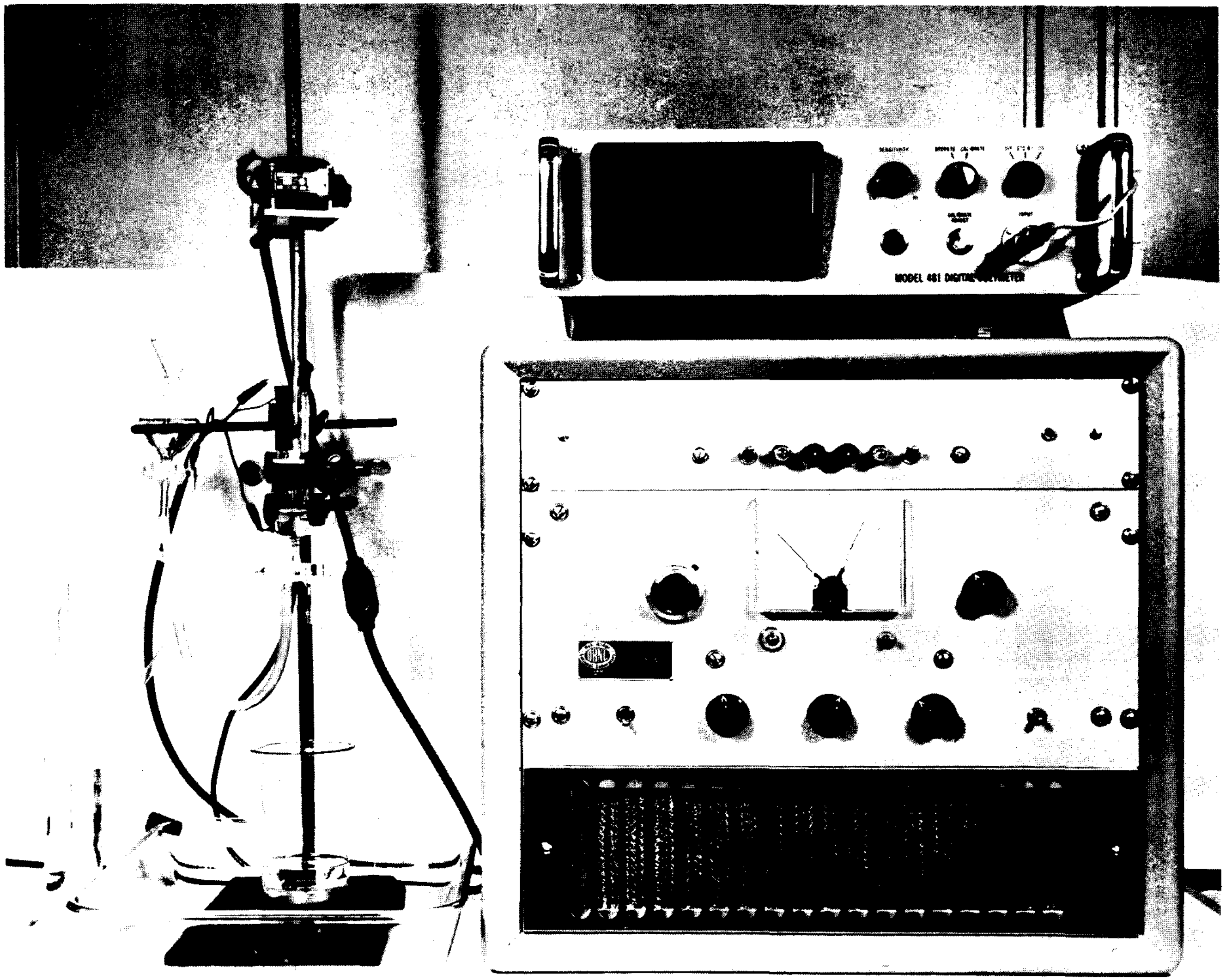


Figure 1

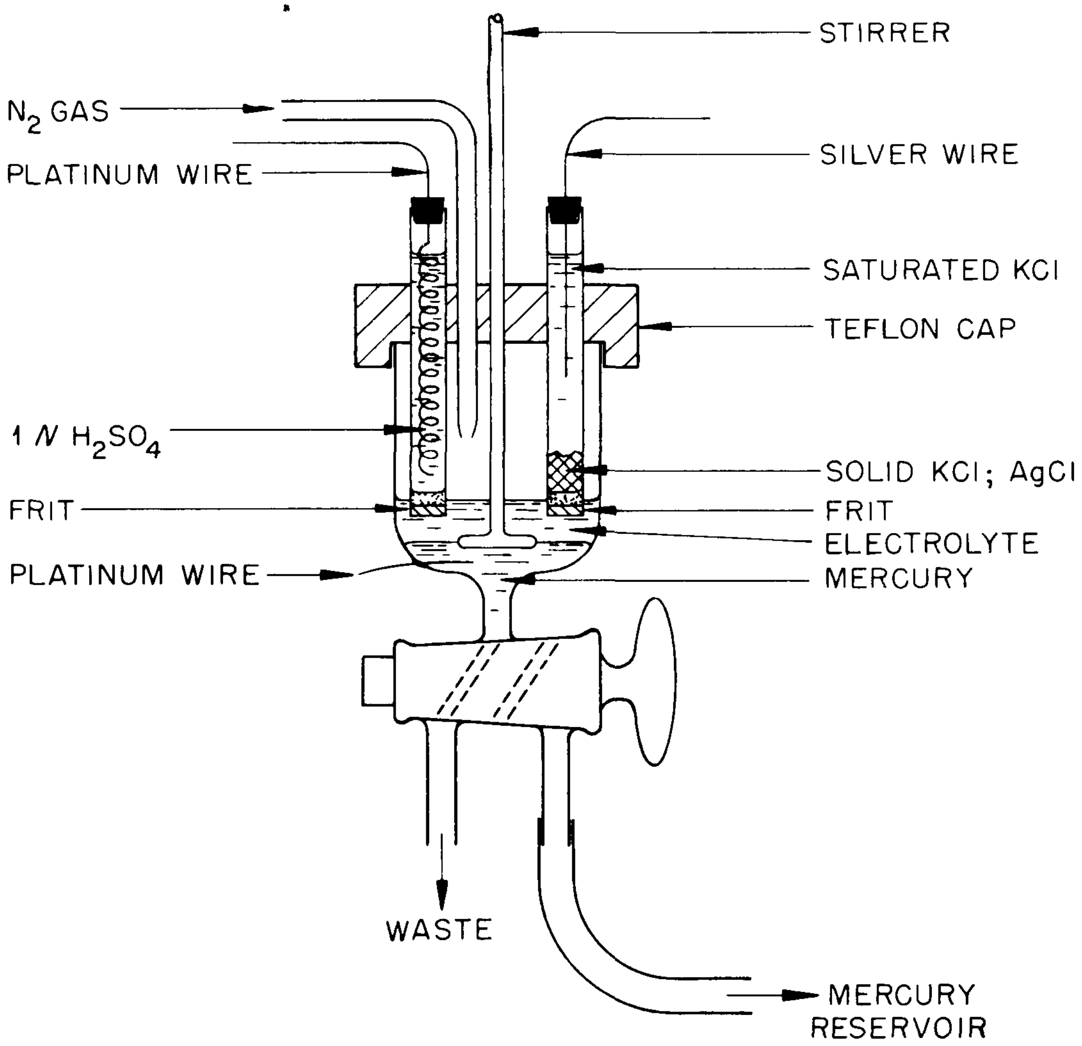


Fig. 2—Diagram of coulometric titration cell.

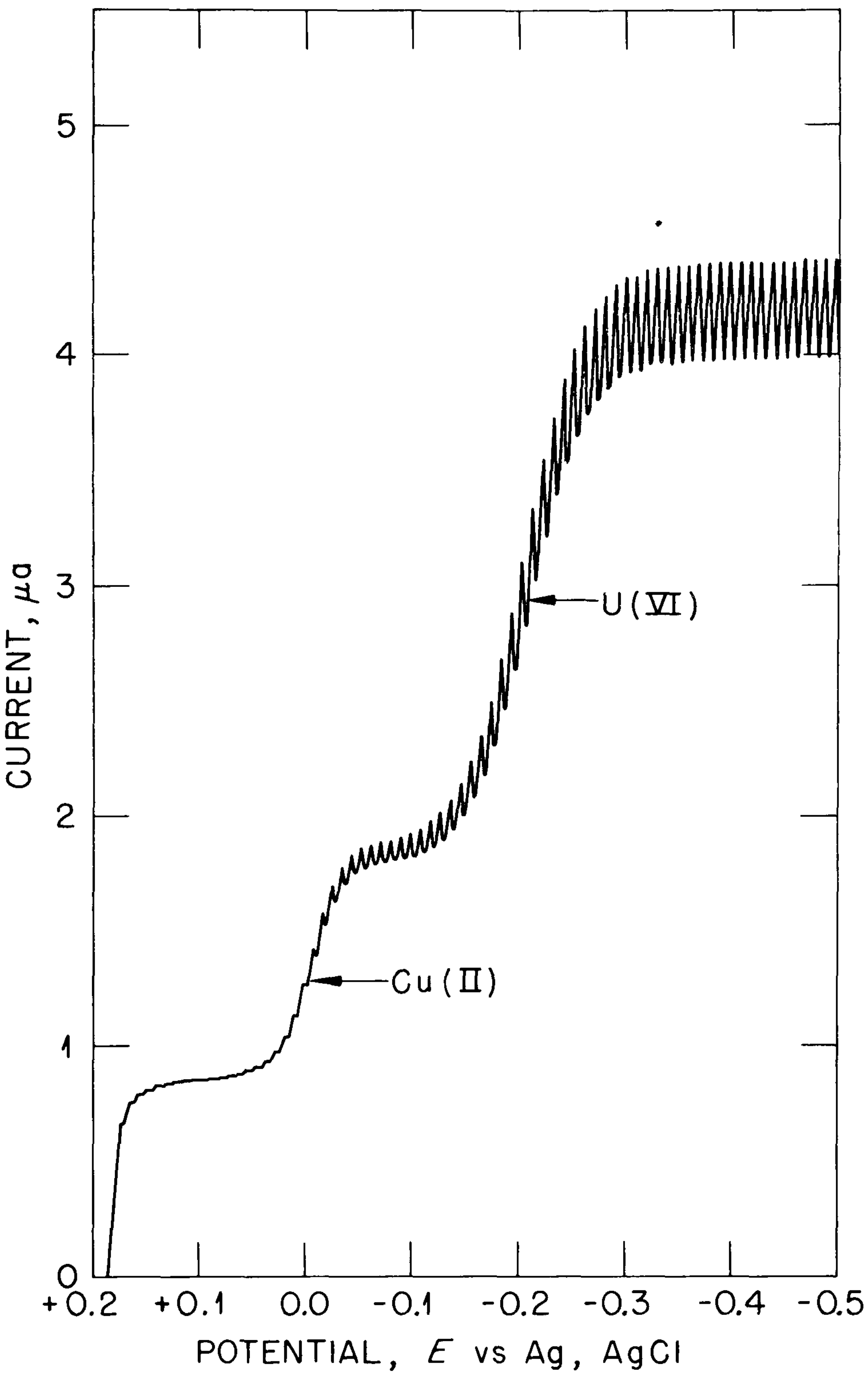
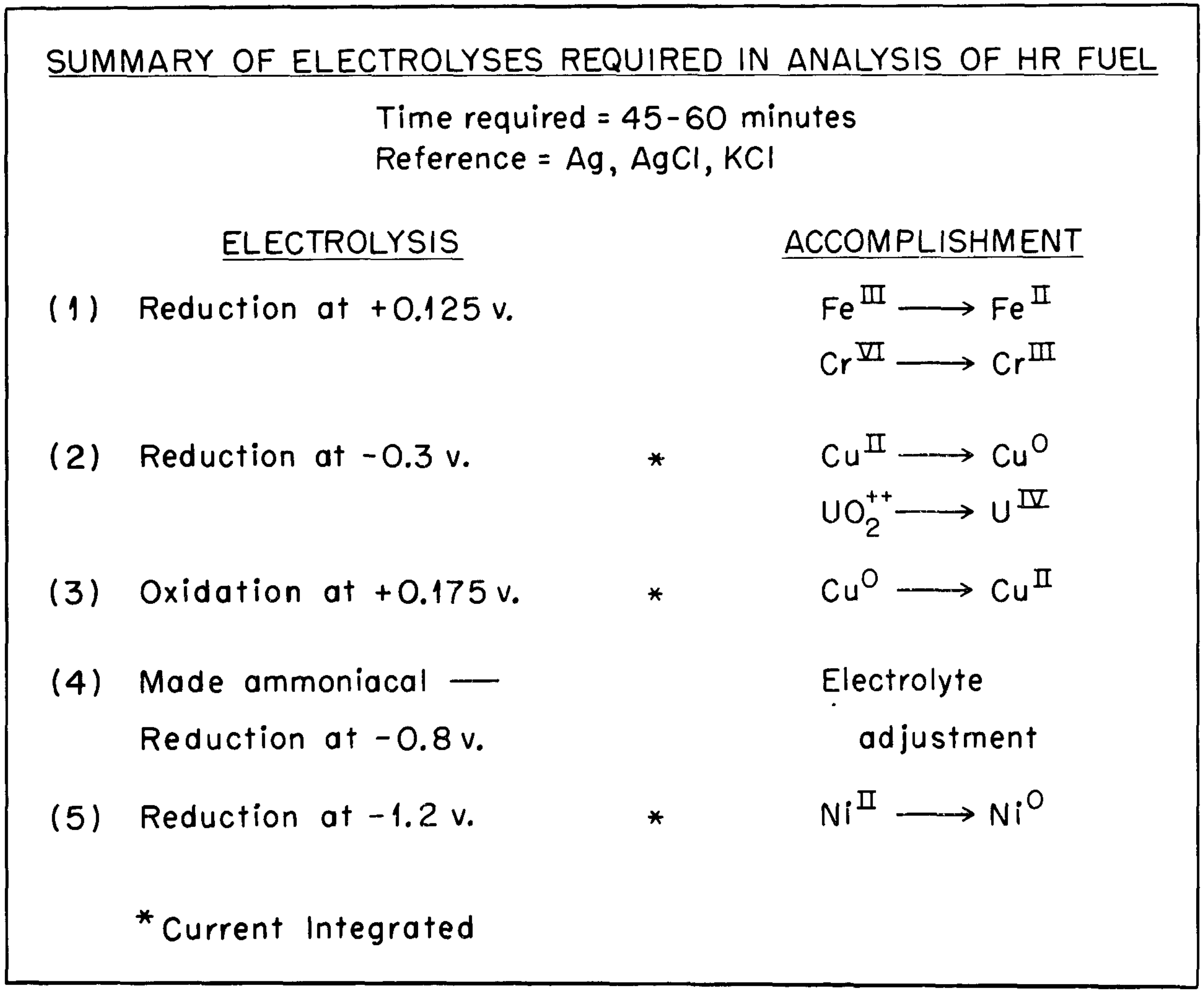


Fig. 3—Polarographic behavior of UO_2^{++} and Cu^{++} in N H_2SO_4. (0.1 mg U(VI)/ml; 0.01 mg Cu(II)/ml.)



SUMMARY OF ELECTROLYSES REQUIRED IN ANALYSIS OF HR FUEL

Time required = 45-60 minutes
Reference = Ag, AgCl, KCl

	ELECTROLYSIS		ACCOMPLISHMENT
(1)	Reduction at +0.125 v.		$Fe^{III} \longrightarrow Fe^{II}$ $Cr^{VI} \longrightarrow Cr^{III}$
(2)	Reduction at -0.3 v.	*	$Cu^{II} \longrightarrow Cu^{0}$ $UO_2^{++} \longrightarrow U^{IV}$
(3)	Oxidation at +0.175 v.	*	$Cu^{0} \longrightarrow Cu^{II}$
(4)	Made ammoniacal — Reduction at -0.8 v.		Electrolyte adjustment
(5)	Reduction at -1.2 v.	*	$Ni^{II} \longrightarrow Ni^{0}$

*Current Integrated

Figure 4

TABLE 1—RESULTS OBTAINED BY CONTROLLED-POTENTIAL COULOMETRIC TITRATION OF Cu^{++} AND UO_2^{++} USING TOTAL REDUCTION AND SELECTIVE OXIDATION

Readout voltages		Difference voltage	Copper, mg		Uranium, mg	
Reduction UO_2^{++} plus Cu^{++}	Oxidation Cu (Hg)	UO_2^{++}	Present	Found	Present	Found
0.0735	0.0735	0	0.966	0.093	0	0
0.1170	0	0.1170	0	0	5.750	5.752
0.1904	0.0735	0.1169	0.966	0.963	5.750	5.749
0.1542	0.0370	0.1172	0.483	0.484	5.750	5.760
0.1244	0.0072	0.1172	0.097	0.095	5.750	5.762
0.3078	0.0735	0.2343	0.966	0.963	11.500	11.516
0.1030	0.0736	0.0294	0.966	0.964	1.438	1.447

TABLE 2—CONTROL DATA (RUN 17, HRT, JUNE-AUG. 1958)

Determination	Number of determinations	True value, mg/ml	Average value	Standard deviation, %	Method of analysis
U	170	10.95	10.99	1.0	Coulometric titration
Cu	170	0.325	0.334	4.0	Coulometric titration
Ni	51	0.120	0.120	8.0	Polarographic
SO_4	55	7.65	7.59	1.0	Photometric titration
XSH^+	29	0.053N	0.050N	6.0	Potentiometric titration

TABLE 3—PRECISION ESTIMATES
(Uranium in dissolver solutions by amine extraction in coulometric titration)

General composition	Origin	Relative standard deviation, %
U, Pu, F.P., HNO_3 (Al, Hg)	Purex Zirflex Zircex Sulfex TBP–25	0.2
U, Pu, S.S., F.P., HNO_3, HCl	Darex	0.4
U, S.S., F.P., HNO_3, H_2SO_4	SIR	0.4
U, Zr, F.P., Al, HF, HNO_3	STR	0.3
Th, U, F.P., Al, HNO_3	Thorex	0.6

fuel. The results were as precise and accurate as on non-radioactive solutions, i.e., about 0.2 percent. Some of the actual HRT fuel was analyzed and then standard additions of uranium were made and the solutions reanalyzed. Essentially no differences were found in the results.

Some dissolver solutions will contain large quantities of iron, nickel, and chromium such as the stainless steel fuels. Other fuels will contain large quantities of zirconium such as the STR type fuel. It would be almost impossible to directly titrate some of these dissolver solutions for uranium. Therefore several solvent extraction procedures have been examined for the separation of uranium from the large amount of other metals. This would be a very selective method of determining uranium since any small amount of iron, chromium, nickel, or zirconium carried through the extraction procedure would not interfere because of the selective electrolysis voltage during the subsequent titration.

The outline of the analysis of dissolver solutions by use of triisooctylamine as the uranium complexer for solvent extractions follows: (Time required, 45 minutes; relative standard deviation, 0.2–0.6%.)

1. 5 ml dibasic aluminum nitrate + 5 ml 5% triisooctylamine in xylene + sample containing 2–12 mg U.
2. Stir 5 minutes, discard aqueous.
3. Strip 5 minutes with 5 ml of 0.5 M H_2SO_4, 0.05 M $HClO_4$, drain aqueous into titration vessel.
4. Repeat (3).
5. Reduce uranium (Hg cathode) at –0.35 v. vs Ag, AgCl.

Hexone (methylisobutylketone) also has been used. It was found that an aqueous stripping of the uranium from the organic phase with a mixture of sulfuric acid and perchloric acid could then be titrated directly without chemical treatment. Synthetic nonradioactive dissolver solutions have been analyzed by this procedure and the results are shown in Table 3.

The relative standard deviation has been calculated on the basis of 10 analysis of each type dissolver solution.

Some solvent extraction work has been carried out in the shielded cave and the mechanical problems in this procedure have been solved. Highly radioactive dissolver solutions are unavailable at this time, but on the basis of the experience with the analysis of the extremely radioactive HRT solutions an adequate method for their analysis should be assured. It is believed that a precision and accuracy of better than 1 per cent can be achieved.

In conclusion a mention of some of the preliminary work on the determination of plutonium by controlled-potential coulometric titration should be made. In this case a platinum cathode is used in an electrolyte of 1 M perchloric acid. The titration procedure is as follows:

1. 7 ml 1 M $HClO_4$ + sample containing 0.2–2 mg Pu.
2. Reduce (Pt electrode) at + 0.125 v. vs Hg, Hg_2SO_4.
3. Oxidize (Pt electrode) at + 0.425 v. vs Hg, Hg_2SO_4.

All of the plutonium is reduced to the Pu(III) valence state by reduction at +0.125 volts vs Hg_2SO_4 and then the current is integrated for its oxidation at +0.425 volts vs Hg_2SO_4.

A relative standard deviation of approximately 0.1 per cent was obtained for the analysis of 10 aliquots of plutonium solution. One milligram of plutonium was titrated.

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MANAGEMENT POLICY AND PHILOSOPHY PERTAINING TO CONTROLS OVER SOURCE AND SPECIAL NUCLEAR MATERIALS AT GOODYEAR ATOMIC CORPORATION, PORTSMOUTH, OHIO

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On October 13, 1952, the Goodyear Tire and Rubber Company was selected by the Atomic Energy Commission to operate a new gaseous diffusion plant to be constructed near Portsmouth, Ohio, for the production of uranium highly enriched in U^{235}.

The Goodyear Atomic Corporation was organized as the operating contractor of the plant facilities provided by the AEC. Figure 1 shows the plant facilities.

During the early construction period, arrangements were made with the Union Carbide Nuclear Company for the training of key Goodyear personnel at the gaseous diffusion plants located at Paducah, Kentucky, and Oak Ridge, Tennessee. The excellent cooperation received by Goodyear during this period made possible a very practical formulation of policies and philosophies with respect to all aspects of operation of a plant of this nature.

One of the important considerations in defining organizational responsibilities was the establishment of adequate controls over Source and Special Nuclear Materials. It was recognized that, regardless of the organizational segment acquiring this function, sufficient flexibility had to be provided to permit plant-wide control.

Parent Company policy was such that a function of this type could not be placed under the jurisdiction of the organization which has the major portion of uranium in its possession. Consequently, the Uranium Control section was created as a major subdivision of the Technical Division of the Atomic Corporation. Uranium materials in the hands of the Technical

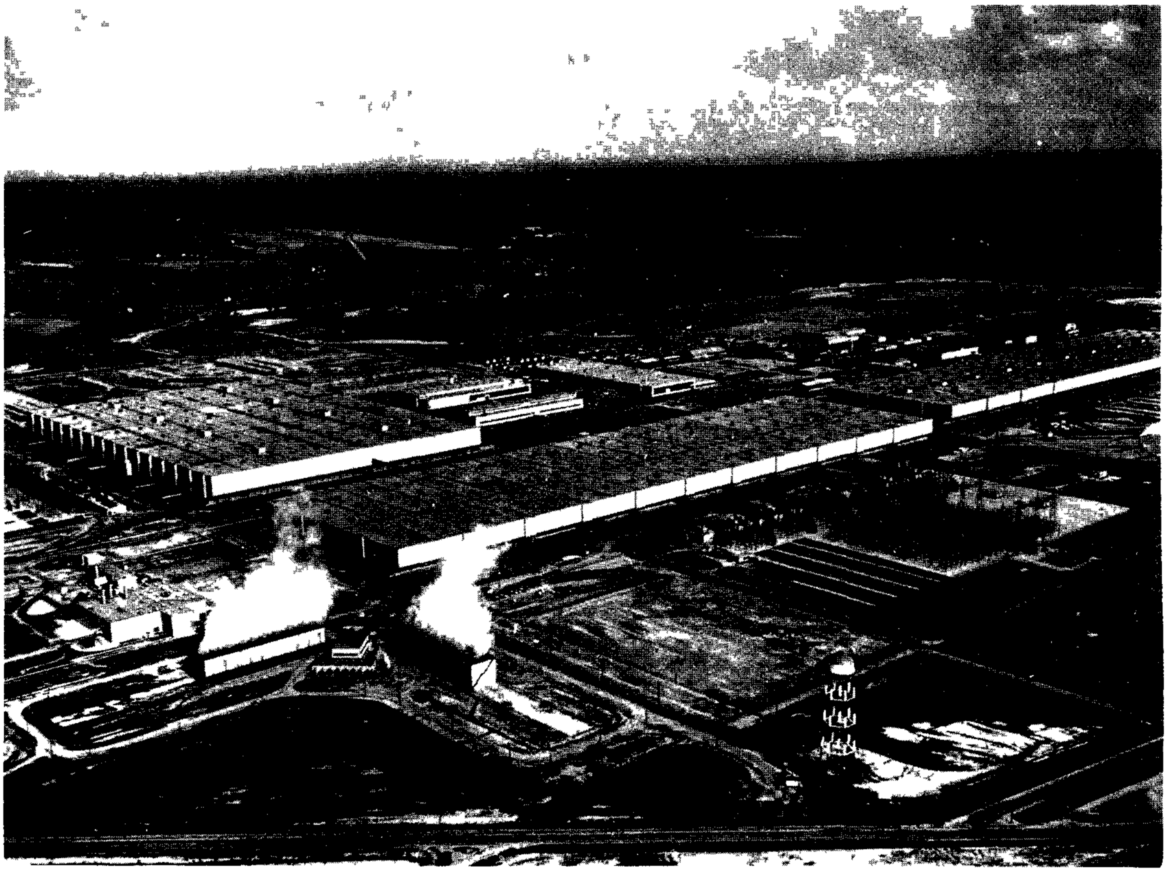

Figure 1

Division represent only 0.5 percent of the total plant inventory. In addition, such organizational structure provides a good line of communication when changes in plant operating parameters, in design of plant equipment, and in laboratory measurements both actual and experimental in nature are made.

Figure 2 gives the functional relationship of the Uranium Control section to other sections of the Technical Division.

POLICY

It is the policy of the Company to abide by the basic principles set forth in Part 7400 of the

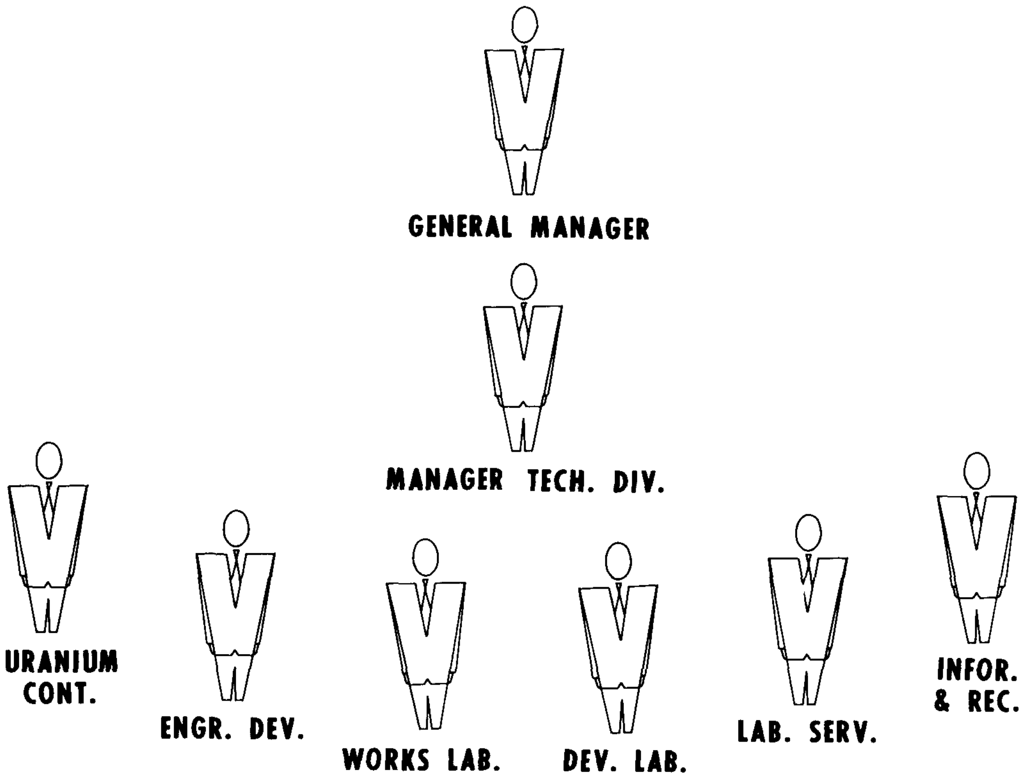

Fig. 2—Goodyear Atomic Corporation organization chart.

AEC Manual. Two of these basic principles are:

1. Source and Special Nuclear Materials supplied the Company by the Atomic Energy Commission will be processed in the most efficient manner possible consistent with economic factors and the strategic importance of such material.
2. The Company will actively support the accountability program in order to provide basic information for controls, conversion, and the accounting for Source and Special Nuclear Materials which have been placed in its possession.

OPERATING PHILOSOPHIES

In order to carry out the intent of this broadly stated policy, it is necessary to fix responsibilities for each organizational unit involved in the production and measurement of uranium-bearing materials.

The Production Division is responsible for the operation of the uranium separation equipment, the decontamination and uranium recovery facilities, and the UF_4 to UF_6 conversion unit. The operation of the associated auxiliaries, such as the electrical switchyards and the sanitary and recirculating water systems, is also included among the operational functions of the Production Division. Since most of the SS Materials are in the possession of the Production Division, it is important that the Uranium Control section be informed of any changes, modifications or experiments being made in the production systems that could cause erroneous information to be reported in the monthly SS Accountability Reports.

The use of prescribed forms covering movements, inventories, and production data, as they affect SS Materials measurements, provides the required information for the operation of the SS Materials Accounting System. Conserving SS Materials and preventing their unauthorized diversion or withdrawal are prime responsibilities of the Production Division. The preparation of permanent or temporary operating specifications covering production goes a long way toward providing current information that may or may not affect uranium measurements. Approved copies of all operating specifications are provided for the Uranium Control section. In some cases, specifications are originated and/or approved by the Uranium Control section prior to their adoption.

The engineering departments whose functions are to design, change, and modify equipment and to conduct tests or experiments related to production systems, are organizational units under the jurisdiction of the Technical Division. This serves to provide both written and verbal information to the Uranium Control section in order to assure that material losses are held to a minimum and that adequate measurements can be made or maintained prior to the making of changes in the production systems.

A very important group is the Works Laboratory whose prime responsibility involves the analytical measurement of uranium and uranium-235. They supply, clean, and move all sample containers to and from production areas using properly executed Material Transfer forms. Standards are prepared to which accountability samples are referred.

An "intralab" control program, in which disguised samples are repeatedly submitted for analysis, provides data for statistical evaluation of the precision of the uranium measurements. In all material flows, the exchange of samples between the shipper and receiver supplies additional data concerning sampling and analytical results for control purposes.

Let us examine the organization of the Uranium Control Subdivision as it is shown in Fig. 3. The duties attached to my office are to request all SS Materials required for use by the Company; to act as a liaison between the Company, the AEC, licensees, and other AEC contractors in supplying SS Materials for their use; and to serve as the accountability representative for the Company. It can readily be seen that the Uranium Control Subdivision is

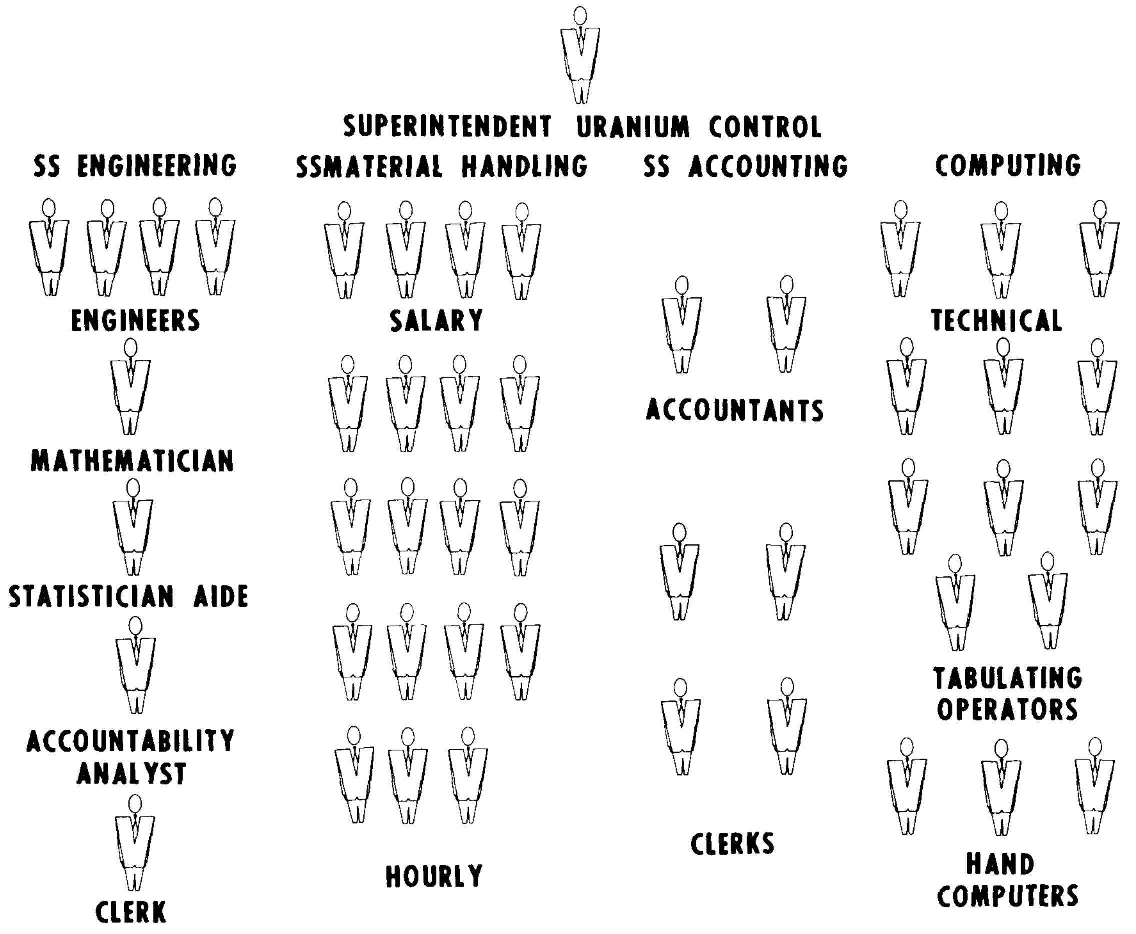

Fig. 3—Uranium Control Subdivision organization chart.

organized in a functional manner and is similar to the larger divisions of the Corporation. We have an Engineering Branch, a Transportation Branch, an Accounting Branch, and a Control Branch.

Now let us examine the responsibilities of each of the subsections individually. One of the most important responsibilities of the SS Engineering Branch is to establish controls and procedures to be used between contractors for interplant flows. In addition, they are to prepare and issue instructions for the taking of periodic inventories of SS Materials. I would like to say that this is one responsibility that none of the men relish since none of them have had the opportunity to attend a New Year's Eve party since this organization was set up. This situation is understandable since one of their responsibilities is to be present, observe, and deal directly with designated supervisory operating personnel at official inventory times. You are all familiar with the fact that inventories are taken at a gaseous diffusion plant by supplying data for calculations based on the common gas laws. In order to compile the data, the engineers have to determine and verify the volume represented in process equipment which is significant in the inventory calculations.

Another important inventory responsibility is to determine that all instruments and scales have been accurately calibrated so that all measurements pertaining to SS Materials may be correctly taken. Two responsibilities which are tied very closely together are those concerning releases or spills and those concerning the investigation of discard limits, consistent with economic considerations, for uranium-bearing materials. In order to carry out these two responsibilities, it is necessary to investigate and determine if any changes or proposed changes in design, any modifications, or any experiments involved in operations using SS Materials would cause erroneous information to be reported in monthly accounting statements.

The SS Materials Handling Department acts as dispatcher and receiver of all material entering and leaving an area. It is responsible for the storage of all SS Materials and must provide, number, and tare weigh all containers for SS Materials. This section also establishes the official weight of all SS Materials containers used for accountability purposes.

The SS Accounting Department has provided an accounting system, complete with records and controls, that will meet the basic requirements of AEC regulations. This includes the design, issuance, and accounting for all serially numbered forms used for gathering source information. This information, as you know, is supplied on a monthly basis in accordance with AEC regulations. In addition, this section provides the Finance Division with data concerning SS Materials movements and inventories for use in the making of financial statements in various reports.

And last, but not least, is our Control or Computing Department. They are charged with the responsibility of establishing proper mathematical equations and they calculate our dynamic inventories. They also supply, on a plant-wide basis, computing services of a high mathematical order, utilizing high-speed data-processing machines or desk calculators.

The Uranium Control Subdivision is responsible for assisting all supervision in problems related to SS Materials accountability throughout the plant.

It can readily be seen that the possibility of SS Materials arriving on the plantsite with the Corporation not having definite knowledge concerning the supplier and the intended user is rather remote.

I would like at this time to amplify on one of our Company's responsibilities. We are required to maintain constant surveillance to ascertain if too much or too little effort is being expended in accountability matters. Over the past two-year period, significant savings have been realized through the statistical evaluation of the various sampling programs. This evaluation has led to a reduction in the number of samples removed and the associated laboratory analyses required. Increased filling limits for standard containers and more accurate measurement of SS Materials in their most concentrated forms, both liquid and solid, have also contributed to lower operating costs.

Constant changes in economic factors and in the production and shipment of SS Materials for various and sundry uses require sound but flexible control over uranium quantities. Although the Goodyear Atomic Corporation is something of a newcomer in the field of atomic energy, we feel that the procedures and organizational structure established for SS Materials control have been adequate.

ISOTOPIC ANALYSES BY USE OF TRANSISTORIZED SCINTILLATION SPECTROMETER

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The gamma ray spectrum has been of interest to science for many years, but intensive evaluation of the various spectra was limited to the instruments and equipment of the time. As new instruments were developed, scientists began to learn more about the gamma ray spectrum as related to specific isotopes.

In recent years, gamma ray spectroscopy has come of interest as an analytical tool.

This paper will discuss: (1) the uses of the instrument and analytical methods as a production control or quality control procedure; (2) a new portable transistor scintillation spectrometer; (3) methods of determining U-235 content in various fuel elements foils, etc.

THE USE OF THE INSTRUMENT AND ANALYTICAL METHODS AS PRODUCTION AND QUALITY CONTROL PROCEDURES

An accurate, fast, nondestructive method of uranium analysis is highly desirable during the fabrication and production of reactor fuels and core assemblies. Manual Chapter 7401 states, "Each receiver shall independently measure SS materials received .... The measurement method will consist of weighing, sampling, and analyzing each unit, batch, or container of material, and where appropriate, determining the isotopic composition." This not only makes the determination of the U-235 content of transferred materials desirable, but mandatory.

The methods described in this paper are actually quality control procedures via applied physics.

Just as any appliance or consumer goods manufacturer must utilize good industrial control practices in the manufacture of its wares, so should fuel element and reactor core fabricators utilize good materials control practices. The nondestructive uranium U-235 analytical methods described or similar ones could and should be utilized in the production and fabrication of reactor cores and components.

It has been found that the use of nominals and so called fabrication data may vary from the actual U-235 content from 1% to 20%. These figures were obtained from various items supplied to us by contractors using nominals and fabrication data.

It is essential that physics data from critical assemblies for R&D be accurate for it is upon this data that commercial reactor design parameters are based. If the physics data is accurate, then the commercial or production reactor must be within the specifications as prescribed by the physics data.

Since most reactors utilize U-235 as fuel to produce heat for steam or power production, it can be seen that it is essential to know the U-235 content of the reactor and its distribution within the reactor.

Materials control has become increasingly important with better and stiffer competition. Controls have become more important to meet the demands of stockholders and product demands of consumers.

Materials controls leads to a good specification product that will be accepted by the consumer and manufactured at a reasonable cost by and profit to the manufacturer.

As the commercial fabrication and licensee program expands, the buyer will want to know if he is getting what he is paying for. At a 4% use charge, it makes good business sense to know how much U-235 has been received. He will not wish to destroy any fuel elements - he will have to employ an accurate nondestructive method.

Quality control of a product, whether it be a toaster, chemical, auto or reactor fuel element, is cheap product insurance.

TRANSISTOR SCINTILLATION SPECTROMETER

Introduction

An easily transportable, accurate, transistorized gamma ray scintillation spectrometer is in the final development stage. For easy maintenance, all circuit components are mounted on plug-in units. Wherever feasible, these units are printed circuit cards. The entire instrument is composed of two packages. The main unit which holds virtually all the electronic circuitry is 11" x 12" x 17" long and weighs less than 40 pounds. The second unit which houses the detector is 7" x 8" x 14" long and weighs less than 10 pounds. The detector consists of a 2" diameter by 2" long NaI(Tl) crystal, an RCA 6655 photomultiplier tube, and a pre-amplifier. The main unit consists of a linear pulse amplifier, a differential pulse height analyzer, a count rate meter, a scaler, and a high voltage power supply. The instrument operates on 117 volts AC; however, it can easily be adapted to 220 volts AC. Since the instrument is not frequency regulated, it can operate on 50 cycles as well as 60 cycles, which makes it convenient for overseas operation.

Linear Pulse Amplifier

The amplifier consists of two feedback loops. Each loop includes three DC coupled transistors. The high feedback factor in each loop assures good linearity and gain stability of the amplifier. The over-all voltage gain of the amplifier is 250. It has a rise time of less than 0.4 us and a maximum output pulse of 10 volts. The amplifier gain is adjustable from 0-100% in steps of 1%.

Differential Pulse Height Analyzer

The analyzer operates on a "window amplifier" principle. The pulses ride on a continuously adjustable base line. At each base line level, pulses of a certain height will fall in the window. Such pulses will produce output pulses which will be counted by the scaler and count rate meter. The base line can be adjusted from 0-100% of the maximum usable pulse height by means of the "window level" control, a 10-turn potentiometer. The "window width" is variable from 0-10% of the maximum pulse height, also by means of a 10-turn potentiometer. The settings of the dials are linearly related to the amplitude of the input pulses which produce an output. A "window amplifier" with voltage gain of 10 separates the level discriminator from the window discriminators thus further increasing the stability of the analyzer. By means of a switch on the front panel the analyzer can be converted to an integral analyzer. This means that all the pulses above the base line will be counted. To calibrate the analyzer and count rate meter and check out the scaler, a precision pulse generator is incorporated.

Count Rate Meter

The count rate meter consists of a regenerative circuit which deposits a fixed charge on a capacitor for each pulse to be counted. A voltage proportional to the counting rate is produced across the capacitor as the charge leaks off through a high value resistance connected across it. The linearity between voltage and the counting rate is achieved by means of a feedback amplifier. The meter range can be selected from a low range of 100 counts per second full scale to the maximum counting rate of 10,000 counts per second full scale. An output for a strip chart recorder is provided on the back panel.

Scaler

The scaler consists of six interchangeable decades. Each decade includes a blocking oscillator driving a Sylvania 7155 miniature glow transfer tube. The maximum counting rate of the scaler is 20,000 counts per second. The six decades are capable of storing one million counts. The counting time can be controlled manually or by means of an external timer.

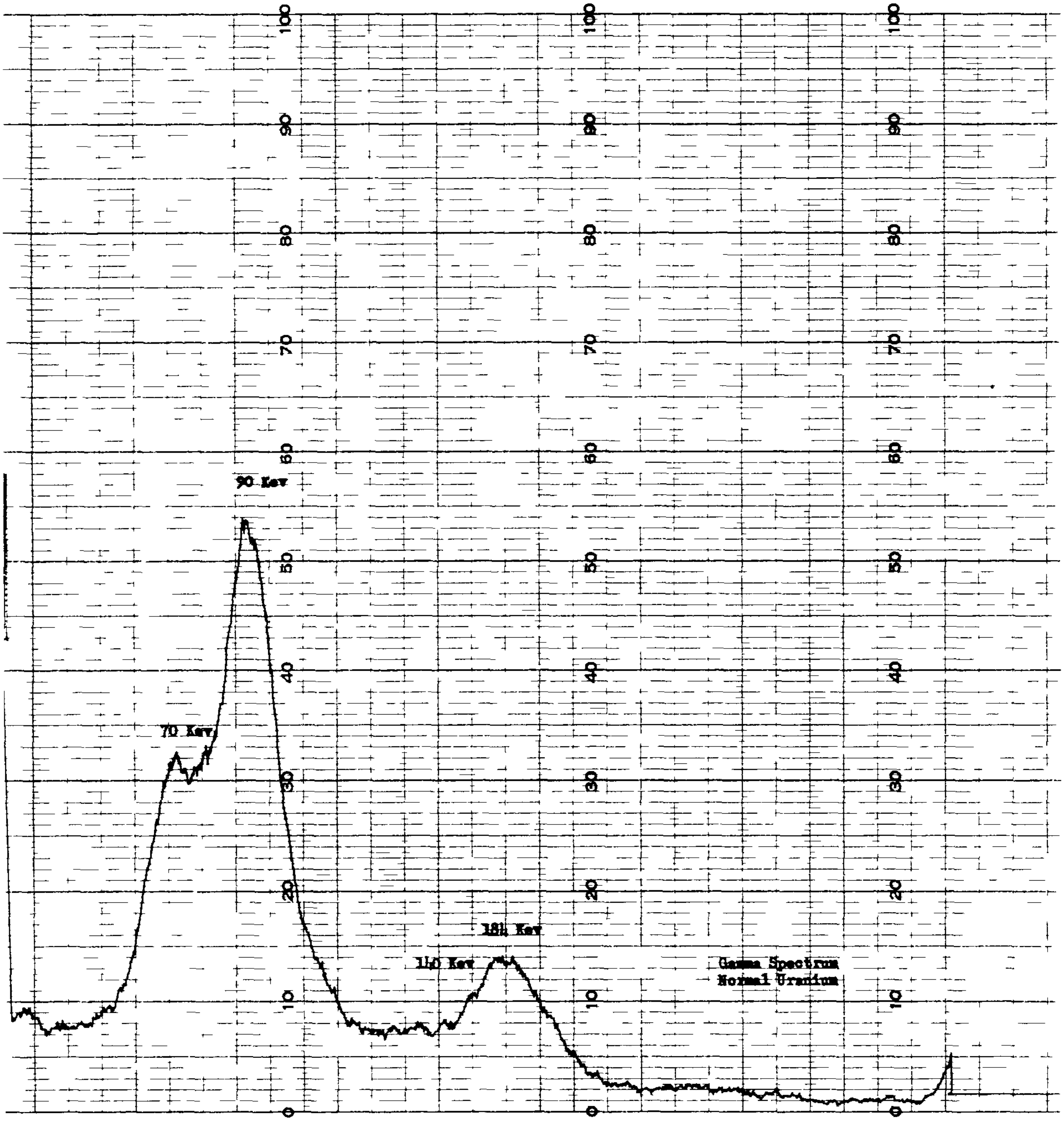
High Voltage Power Supply

This supply provides the negative high voltage needed for the photomultiplier tube. It has a maximum voltage of 1000 volts. Good high voltage stability is achieved by means of double regulation. The voltage is first regulated by means of a shunt regulator consisting of ten 100 volt zener diodes. It is then subjected to further regulation consisting of a transistor series regulator with a high gain amplifier. The high voltage is adjustable from 600 to 1000 volts in steps of 100 volts. Being portable, this instrument can be used in more than one location, the spectrometer need not be lost to the Physics lab.

This instrument is approximately one-tenth the weight of commercially available single channel scintillation spectrometers consisting of relay rack mounted high voltage supply, linear amplifier, pulse height analyzer, scaler, count-rate meter, etc., and consumes about one-twelfth the space.

METHODS AND TECHNIQUES OF U-235 ANALYSES BY GAMMA RAY COUNTING

Using a scintillation spectrometer and scanning the gamma ray spectrum emitted by uranium, the energies are found to be centered around two peaks, one at 90 Kev and one at 184 Kev. Figure 1 shows the spectrum of normal uranium. Figure 2 shows the spectrum of 93% enriched uranium. Figure 3 shows a comparison of the normal uranium spectrum to the spectrum of 93% enriched uranium using the log scale output of the count rate meter. The weight of uranium in both the normal and enriched spectrums were the same.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90 Kev
70 Kev
184 Kev
140 Kev
Gamma Spectrum
Normal Uranium

Figure 1

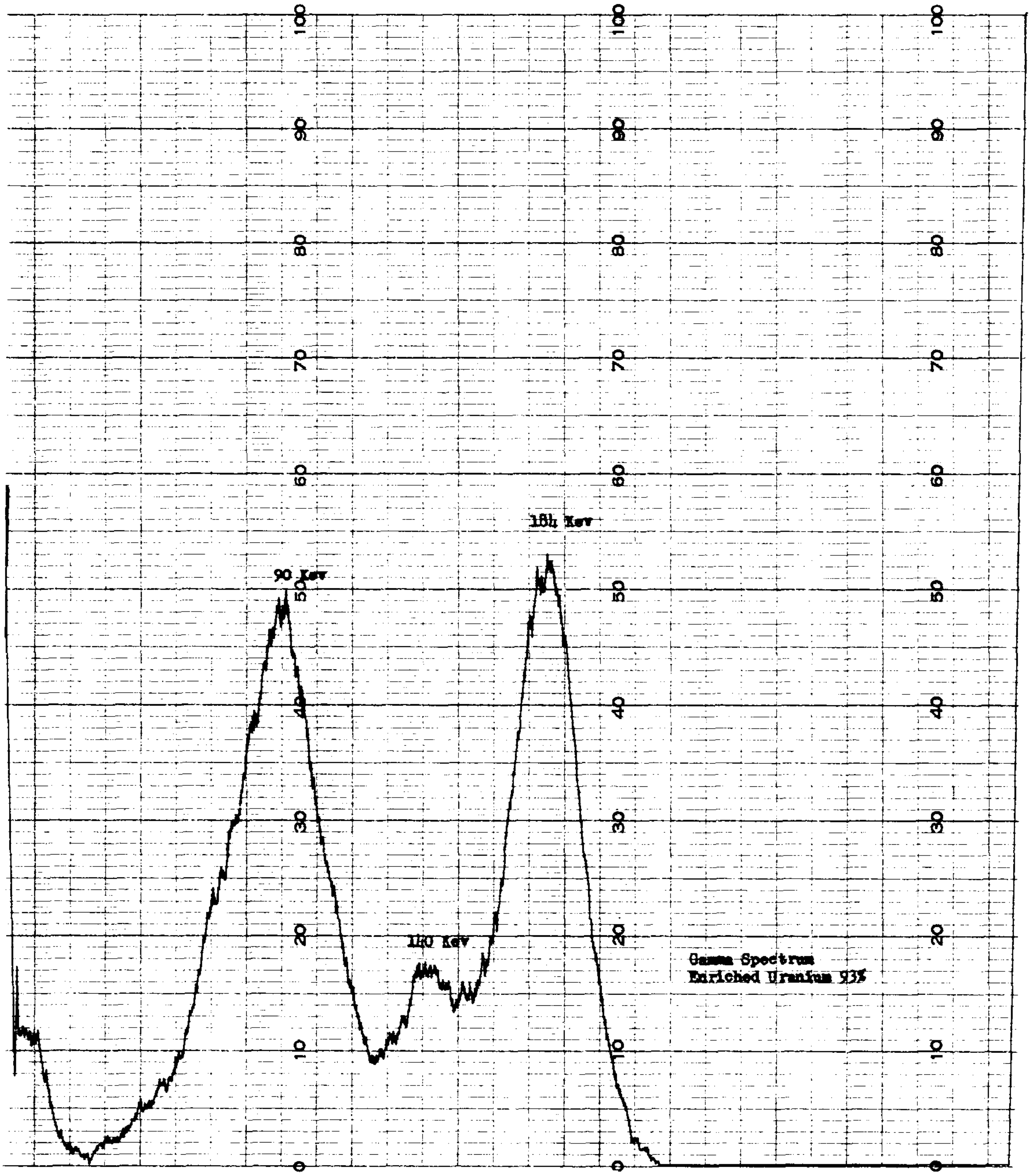
90 Kev
184 Kev
140 Kev
Gamma Spectrum
Enriched Uranium 93%
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Figure 2

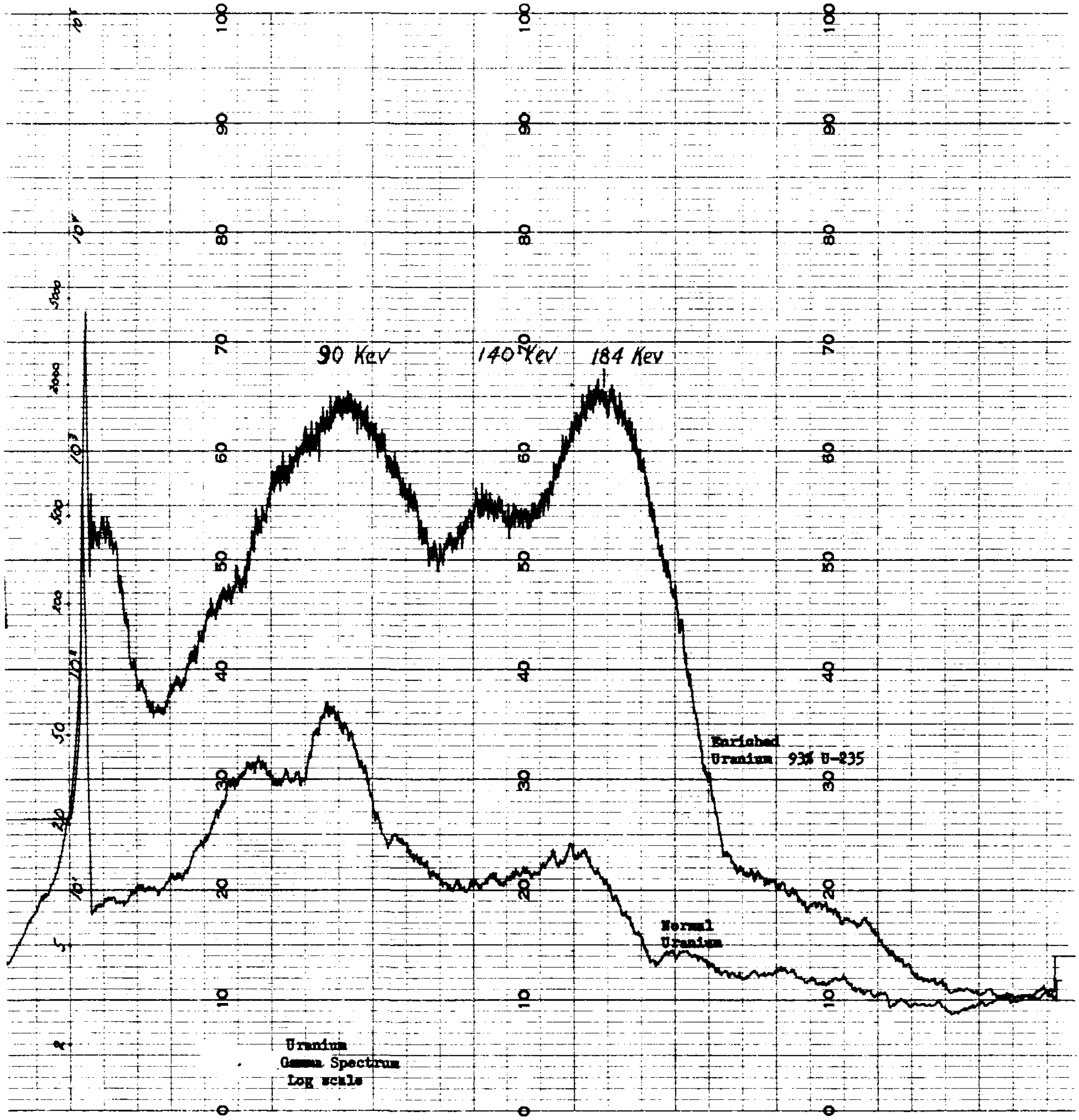
90 Kev
140 Kev
184 Kev
Enriched
Uranium 93% U-235
Normal
Uranium
Uranium
Gamma Spectrum
Log scale
10^5
10^4
5000
2000
10^3
500
200
10^2
50
20
10
5
2
100
90
80
70
60
50
40
30
20
10
0

Figure 3

The 184 Kev gamma rays result primarily from the decay of U-235. The 90 Kev gammas result from U-235 decay and daughter products of U-238. There is also some indication that this peak is partly a result of K X-rays from ionization of atoms by alpha particles. Since the 184 Kev gammas are peculiar to U-235 and this peak is due primarily to gamma energies from U-235, it was chosen as the reference for our analytical method.

In order to determine the U-235 content of an unknown flat fuel element, the following procedure can be used. The unknown is compared to a standard which is made of 1 mil. enriched uranium foil of known weight and uranium content. One mil. foil is chosen because the self absorption coefficient is nearly equal to one for this thickness. $\frac{1-e^{-ut}}{ut}=1$ where (u) is the linear absorption coefficient of the foil and (t) is the thickness of the foil. By keeping our counting rates low, we were able to disregard any corrections for the dead time of our instruments. Background corrections, however, were made.

The geometry is held constant by using a lead collimator. The collimator is made so that the samples being counted are partly shielded, insuring that the same area of the unknown and standard are being counted. The product of ut is determined for the unknown by counting it with the standard foil underneath it and using the equation for linear absorption $I = I_0 e^{-ut}$

To correct for self absorption, the counting rate for the unknown is then put into the equation $M_0 = M \frac{ut}{1-e^{-ut}}$. In this case (u) is the absorption of both the foil and unknown. M_0 is the number of gamma rays that would have been counted if it were not for self absorption, M the observed count rate.

The corrected counting rate can now be compared directly to the counting rate of the standard foil since the counting rate is proportional to the number of atoms present. This gives the U-235 content of the area of the unknown that is in the collimator. In our work the collimator was a 2¼" x 2¼" square hole in ¼" thick lead. By measuring the area of the unknown, the total U-235 can then be determined. If the unknown is not homogenious, it may be scanned to obtain an average counting rate over its length.

A second method, one used by Argonne National Laboratory to determine the U-235 content in ALPR cores can also be used. In this case, preliminary work was done on a number of core blanks. The blanks were counted and two separate cores having the same counting rate were separated out. One core was taken as a standard, the other chemically analyzed to determine the U-235 content. Two other cores with counting rates less than and greater than the standard were chemically analyzed. A plot of counting rate verses the weight of U-235 was made. This curve was linear over a limited range. Therefore, the change in the counting rate was proportional to the change in U-235 content. The counting rate for each core was compared to the counting rate of the standard to obtain the U-235 content of the unknown.

To determine the accuracy of the method, six core blanks were pickled and then counted. These cores showed a loss of U-235 of from 100-300 milligrams which agreed with the weight of uranium lost in the pickling process.

To determine the sensitivity of the equipment and method to changes in uranium content, the following technique was used. A one mil. strip of highly enriched foil weighing approximately 0.8 gram was mounted on the standard core blank and counted. The foil was counted in various positions on top of and below the core. It was found that regardless of where this change in U-235 content occurred, the crystal counted it. The counting rate remained independent of the position of the foil. Therefore the crystal counted the overall core and the counting rate was the overall average of the core. In this case no collimator was used, but the crystal was shielded with lead.

After the preliminary work described above, the actual U-235 content of Argonne Low Power Reactor Cores was determined by counting the 184 Kev gamma from U-235 differentially for five minutes. A 2% "window width" giving a counting rate of about 10,000 counts per second was used to continually check the calibration of the instrument. Every fifth core counted was the standard core determined in the preliminary work.

Both methods, the first one using only a 1 mil. standard, and second, one utilizing a standard the same as the unknowns can be used to determine homogeneity and to determine the total U-235 content in a full fuel element. This is done by scanning the fuel element below the crystal at a constant speed and watching the count rate meter and obtaining the average counting rate for the full element.

In the first method, measurements have been made to within 1% of the actual U-235 content as determined by mass spectrometry.

In the second method, the U-235 content was determined to 0.1 gram in core blanks having a U-235 content of about forth (40) grams.

Acknowledgment is made to:

Harry T. Ryan and Michael G. Strauss of Argonne National Laboratory's Electronics Division for their contribution to this paper and their development work on the scintillation counter.

Warren McGonnagle and Ronald Perry of Argonne National Laboratory's Metallurgy Division for their contribution to this paper and allowing the author to work in their laboratory on the development of nondestructive analytical techniques.

A FAST AND SIMPLE QUALITATIVE METHOD OF DIFFERENTIATING ENRICHED AND NORMAL URANIUM BY GAMMA-RAY SPECTROMETRY

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In the fabrication of nuclear fuel elements, the manufacturer is confronted with the problem of maintaining accurate separation and control of natural from enriched uranium. The methods of separation and control are well known and include precautionary measures for storage, records, and material handling. In addition, the AEC also conducts annual surveys of installations to assure that this control is maintained. It would appear that the possibility of a mistake is remote, but handling of large quantities of SS material by many individuals, will occasionally introduce doubt as to whether certain material contains enriched or normal uranium. When this condition arises, the doubt can be resolved by observing the gamma-ray spectrum of the questionable material.

Gamma spectroscopy is a well known technique for identifying certain elements that characteristically emit gamma rays. The detection device shown in Figure 1 is known as a spectrometer, and includes the following components: a scintillation head consisting of a crystal optically coupled to a photomultiplier tube, a preamplifier, linear amplifier, single-channel analyzer and recording system.

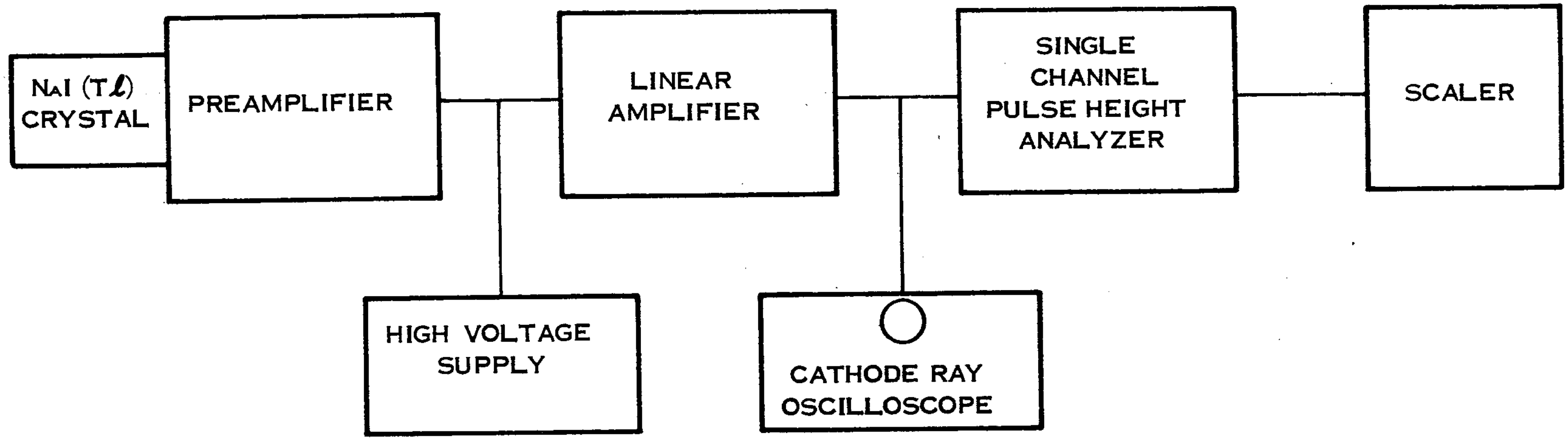

Fig. 1—Block diagram of gamma ray scintillation spectrometer.

A typical pulse energy spectrum of U^{235} (solid line) and of U^{238} (broken line) is shown in Figure 2. It can be seen that the U^{235} spectrum is different from that of natural uranium, and is uniquely defined by two intense photo peaks which appear at 92 and 184 kev. A third peak of lesser intensity appears at 143 kev. The natural uranium spectrum contains only one distinct photopeak which appears at about 90 kev.

Gamma rays of various energies absorbed by the NaI(Tl) scintillation crystal yield negative electrical pulses at the output of the scintillation head. These pulses, after further amplification by the linear amplifier, are fed into an internally triggered oscilloscope with a sweep time of 0.1 microseconds per centimeter. After adjusting the gain of the linear amplifier and selecting a proper rise time, the pulse energy distribution can be spread across the oscilloscope screen. The height of the pulses is approximately proportional to the incident gamma-ray energy.

Figure 3 illustrates the spectra of U^{235} and U^{238} as they appear on the screen of the oscilloscope (Tektronix Model 545).

Here again, as in the case of plotted spectra (Figure 2), the difference between natural and enriched uranium is clearly visible.

In general, chopped stock or melt stock is examined in a well type scintillator; for large pieces of material, a solid scintillator is used.

The method is rapid and simple once the sample has been positioned. Observation of the pattern on the oscilloscope screen will reveal the presence or absence of the characteristic photopeaks.

At the present time, we are investigating methods of determining enrichment over a wide range of values. The integral counting method

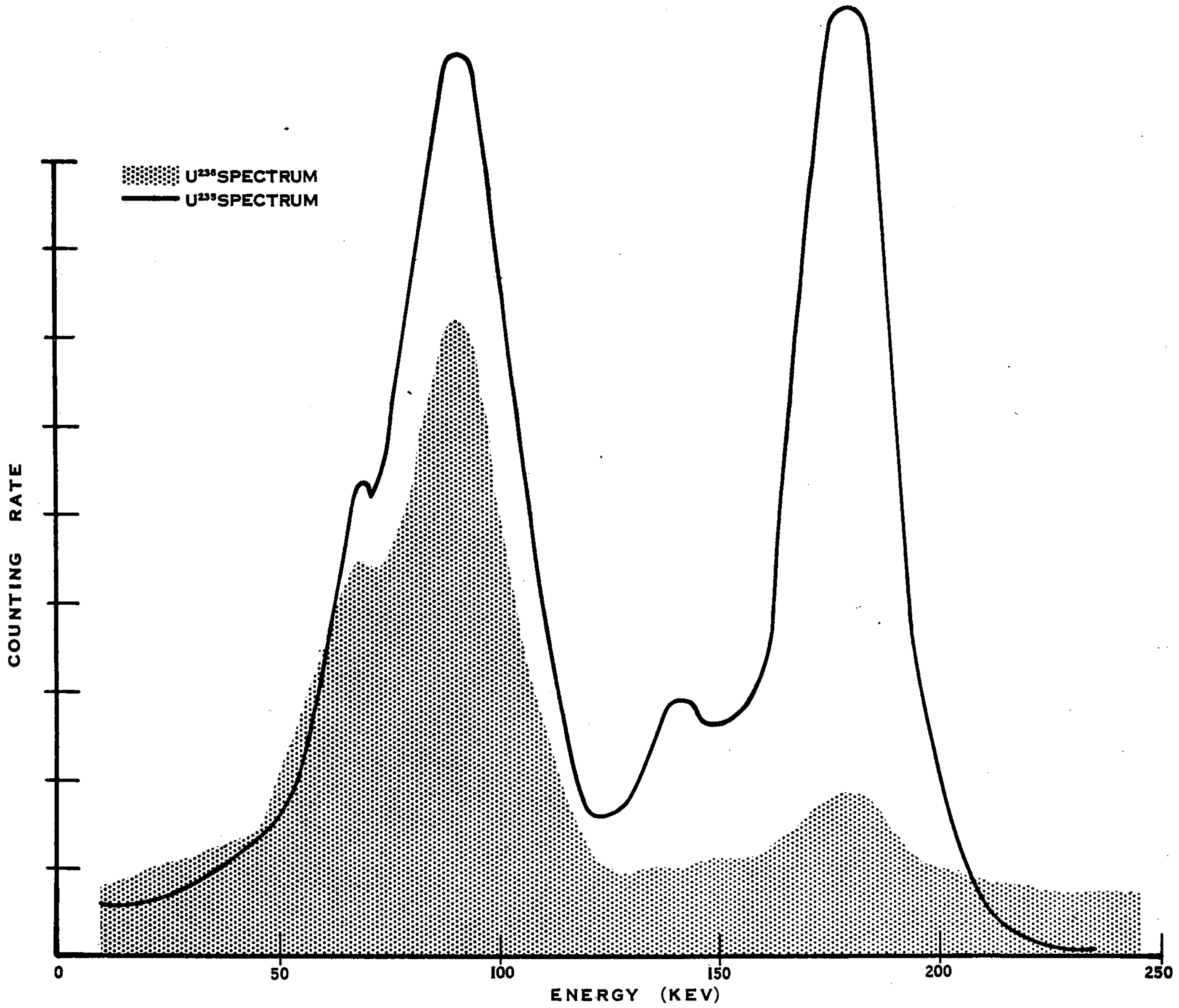

Fig. 2—Typical pulse energy spectrum of U^{235} and U^{238}.

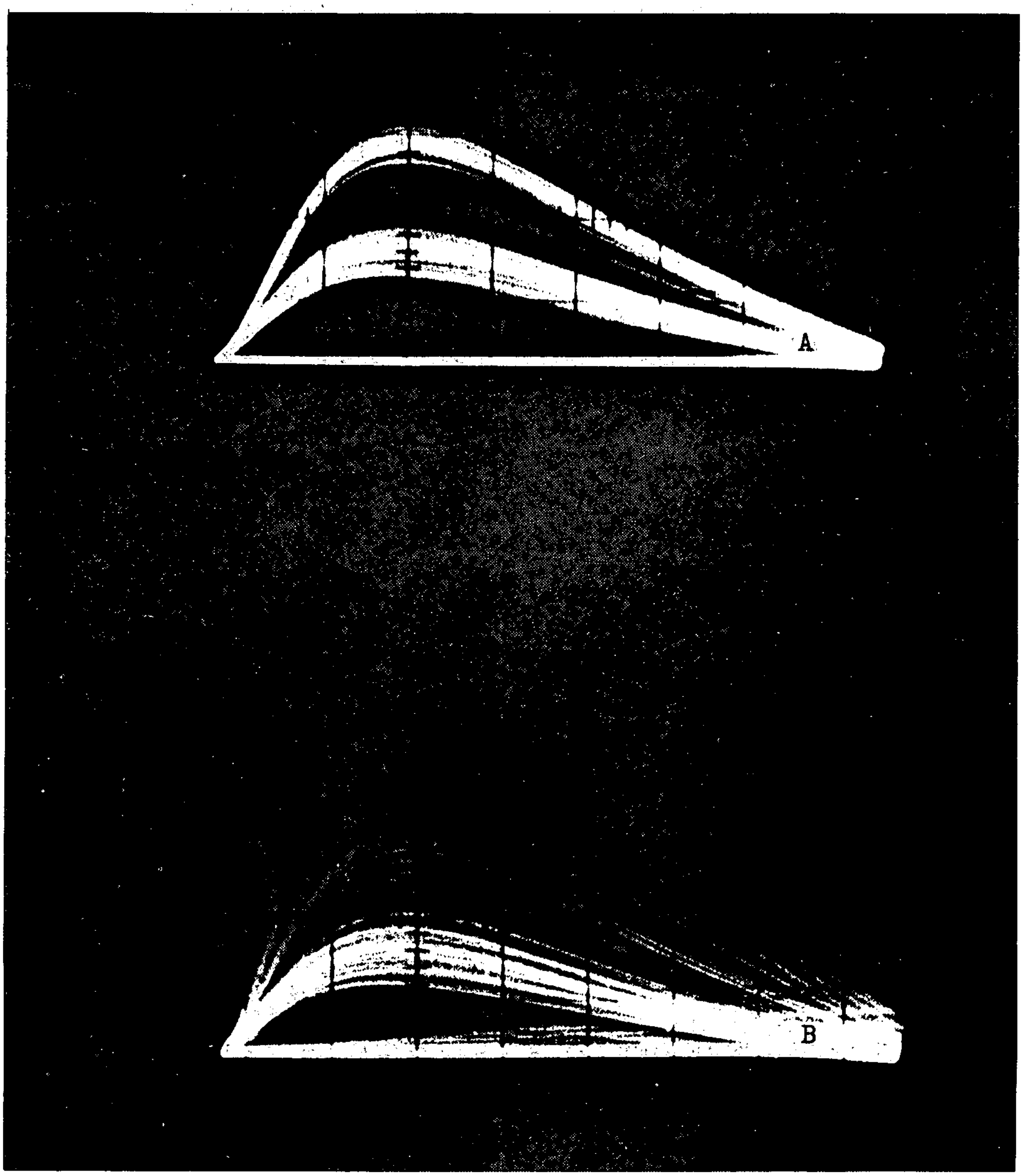

Fig. 3—Oscillograms of the spectra of A. U^{235} and B. U^{238}. The broad bands of intensity clearly show the well-known photoelectric peaks.

has been used, and a series of integral bias curves were obtained for various NBS standard samples from 1.5 - 93%.

Table I compares isotopic values of U^{235} and the measured values.

TABLE I

Sample Number	NBS Value (Atom Per Cent)	Measured Value (Atom Per Cent)
U^{930}	93.33	93.0
U^{900}	90.20	89.8
U^{850}	85.14	86.0
U^{200}	20.00	21.5
U^{050}	5.01	7.0
U^{030}	3.05	5.2
U^{020}	2.04	4.1
U^{015}	1.53	3.4

Data was obtained using a solid 1 1/2" x 1" NaI(Tl) scintillator shielded by a 1" thickness of lead. It is felt that with better geometry, i.e., using a well-type scintillator, the accuracy will be improved, particularly for lower values of enrichment. Also the weight of sample required may then be reduced to an order of 10-20 milligrams total uranium.

Improved geometry may also be obtained using solution techniques. Accurately known weights of uranium and/or uranium alloys can be prepared in known volumes of acid solutions. Reliable solution standards can be synthesized in the laboratory to cover specific ranges of interest. Our work to date has shown that reliable data can be obtained for U^{235} determinations all the way from depleted to highly enriched uranium. It is anticipated that comparable data will shortly be obtained on uranium alloys covering the complete range of possible enrichment values.

METAL SPACE FRAME SHIPPING CONTAINERS

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In the design and manufacture of nuclear cores for power plants by the Bettis Atomic Power Division of Westinghouse, a number of companies are called upon to contribute special talents and facilities. These companies are widely separated and the material with which they sometime work contains uranium.

Problems associated with storing and transporting uranium bearing materials have given us concern, as they no doubt have given concern to some of the people who are present. There are many aspects to these problems such as, radiation safety, criticality safety, shipping regulations, strength of containers to withstand transportation, and last but not least, the cost of containers. This is an extremely large subject and one that I do not wish to discuss in its entirety at this time. I would, however, like to discuss some containers and their application to our operation.

We at the Bettis Atomic Power Division have recently developed shipping containers which have passed all of the tests that we have subjected them to, and information about them may help to solve some of your shipping problems.

I would like to describe these containers to you, not as a design that is entirely complete but rather to stimulate creative thinking which may result in the further development of low cost yet safe containers.

Our container development effort was started as a result of a need for a container 54" x 20" x 20". The conventional welded steel angle constructed (Figure 1) was used. The cost of constructing only the outer metal frame was found to be $415.00. Because of this cost and the length of time required for fabrication, another method was obviously needed.

After a considerable search of possible methods and materials, a standardized galvanized pre-slotted steel angle was suggested by one of our employees. We found that this type of material was readily available and by use of a standard cutter, the members could be cut to size and then fastened together by nuts and bolts. (Figures 2 and 3)

Using the pre-slotted steel angle, we first built a space frame measuring 60" x 24"x 24" (Figure 4). The cost of constructing this unit was $72.00. This size frame compared to the one described in the preceeding paragraph was not exact but the same functions were served.

The cost of fabricating both containers Figure 1 and Figure 4 and the weight difference is illustrated as follows:

Figure 1		Figure 4	
Welded Steel Angle		Pre-slotted Steel Angle	
Cost to Fabricate	Weight	Cost to Fabricate	Weight
$415.00	200 lbs	$72.00	85 lbs

The comparison indicated that if the slotted steel angle space frame would meet strength requirements it would be a satisfactory shipping container. Certainly the equipment needed to make the metal frames was simple as the standard cutter shown in Figure 2 cost $45.00; other than this piece of equipment a standard socket or cresent wrench is all that is required.

To assure ourselves that the metal space frames constructed in this manner would withstand the hazards incident to transporting fissionable material, we made a simulated shipment by placing 300 lbs of steel in the inner container.

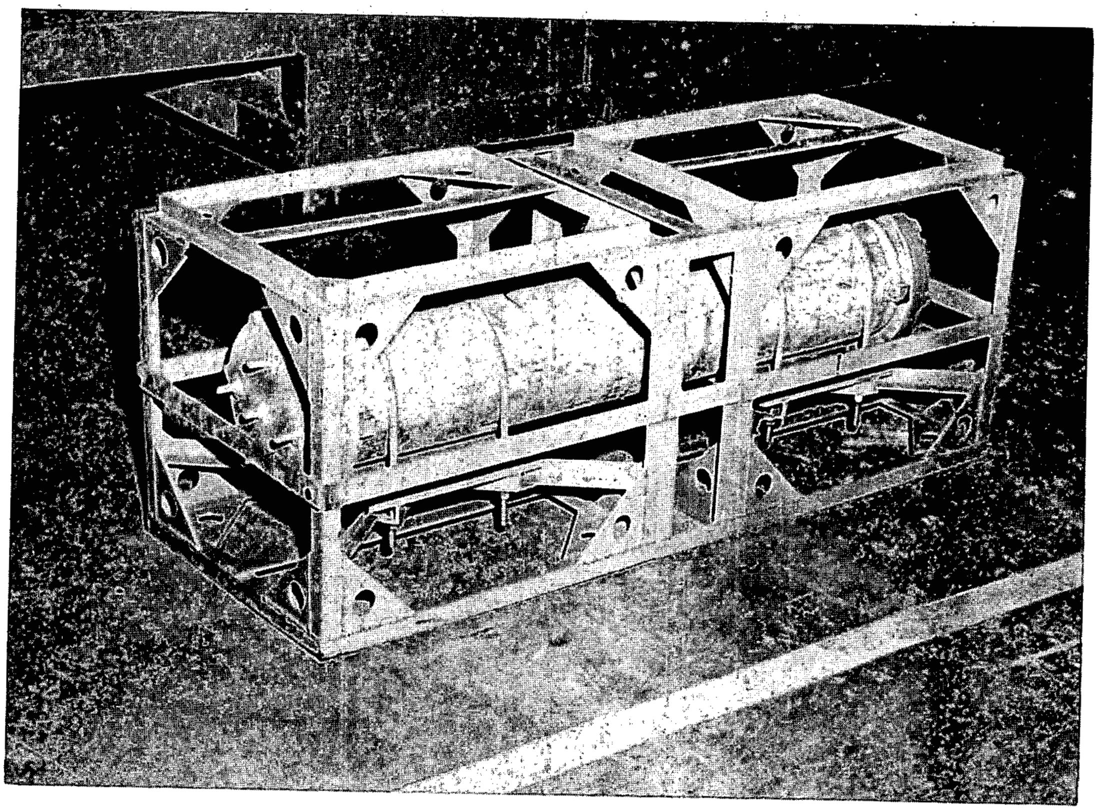

Fig. 1—Metal space frame of welded steel angle, inner container. Eight-in. schedule 40 pipe closed off with end cap.

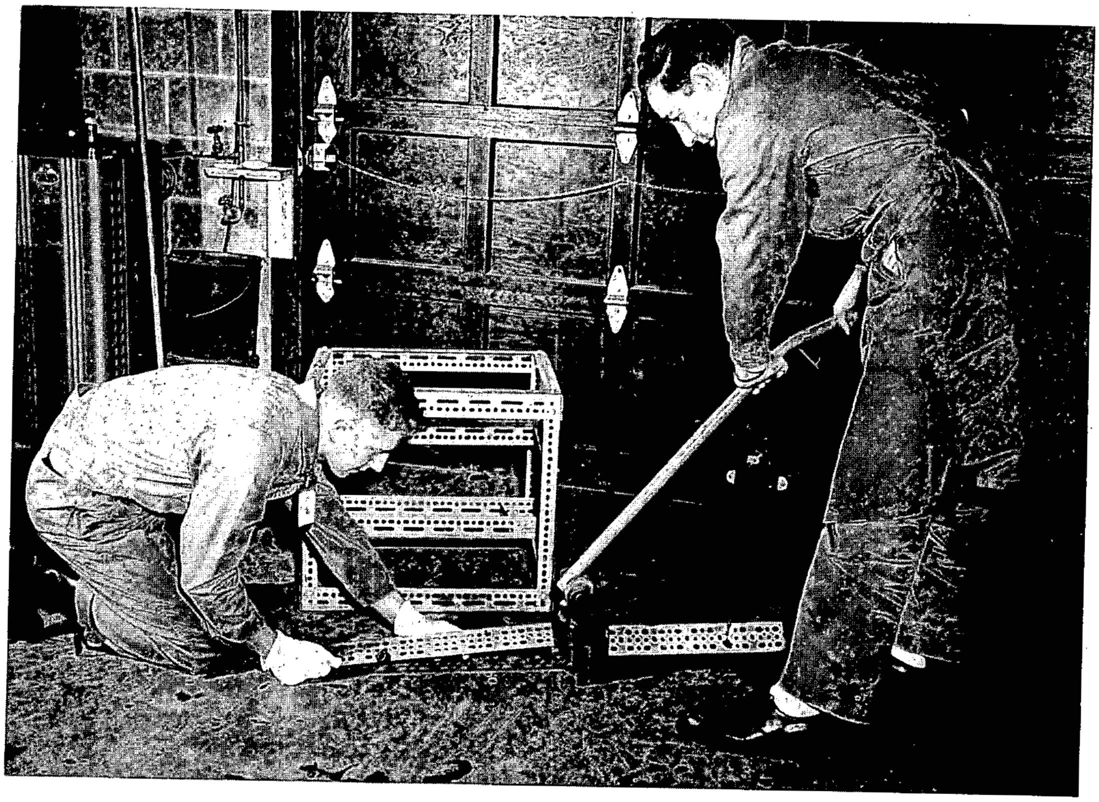

Fig. 2—Cutting slotted steel angle to size.

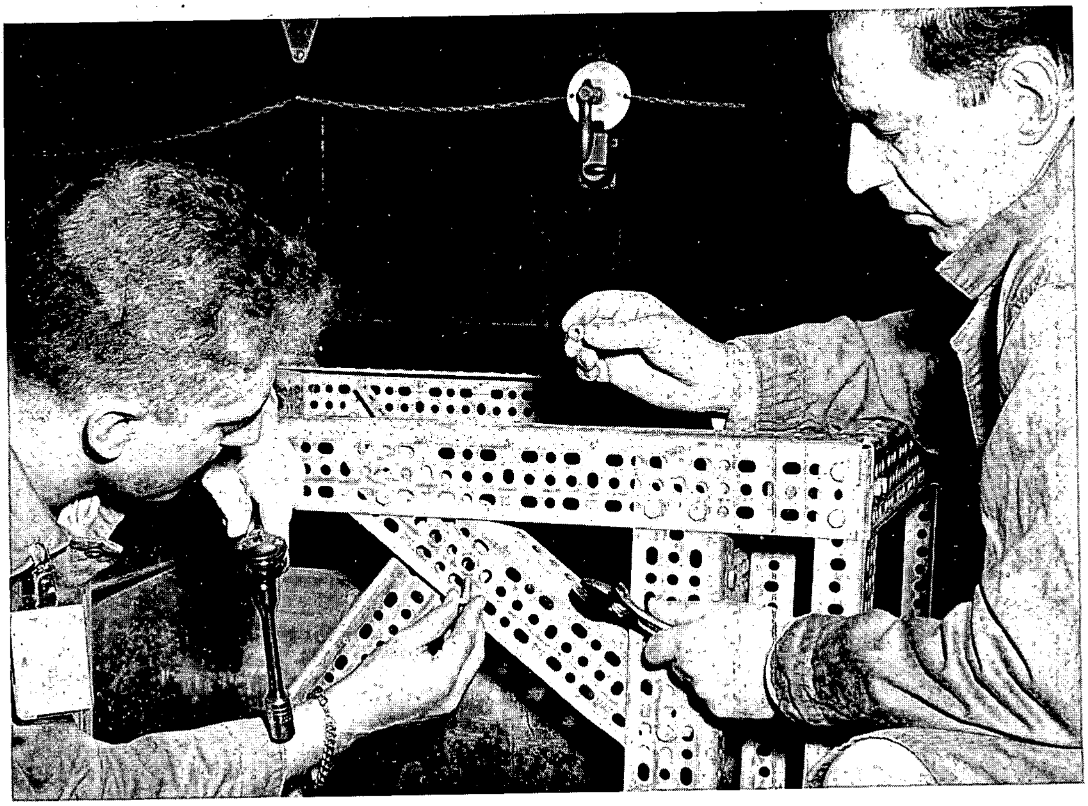

Fig. 3—Assembly of metal space frame.

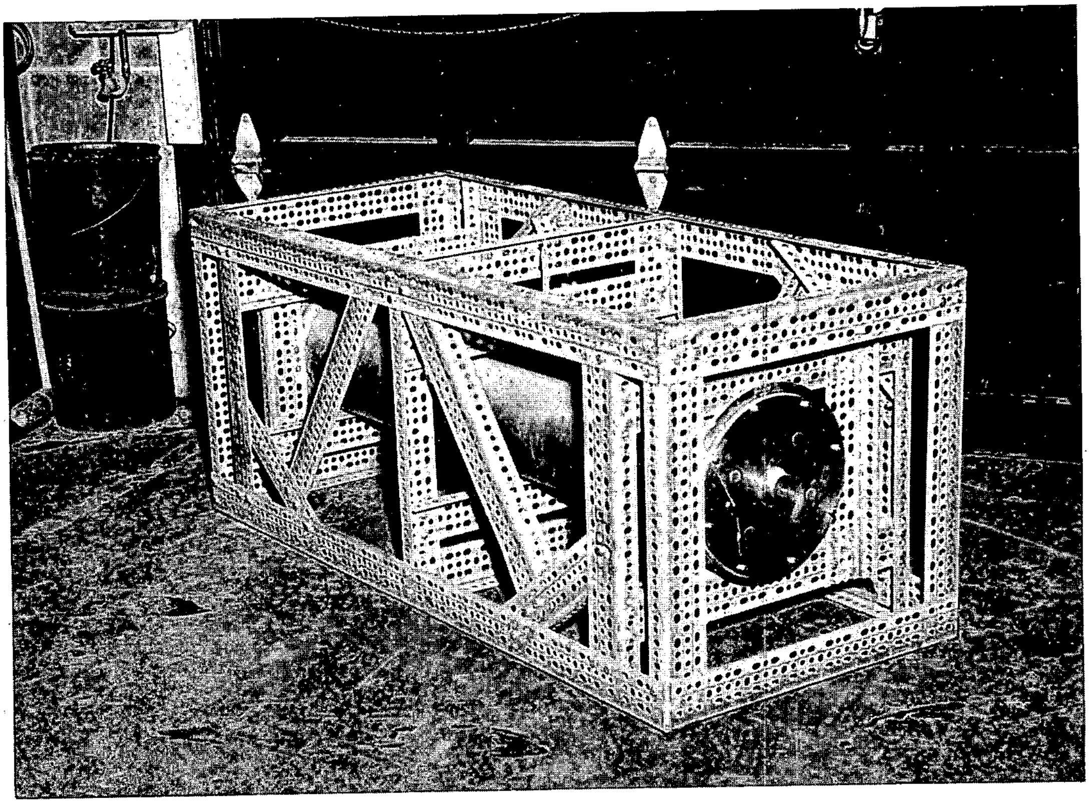

Fig. 4—Metal space frame constructed with slotted steel angle.

The simulated shipment was given an incline impact test (Figure 5) and a vibration test (Figure 6) which subjected the shipment to shocks more severe than would normally be encountered in normal transportation. The tests were equivalent to a round trip between Pittsburgh, Pennsylvania and Chicago, Illinois. The metal frame and its inner container were undamaged and further tests of this type were suspended.

We went a step further and actually dropped the same container from the bed of the truck which was four (4) feet high. In this case we had to replace one (1) slightly bent member. The replacement of the damaged member was easily accomplished. A piece of slotted steel angle was cut and bolted in seven (7) minutes. This you can see could not be accomplished in a welded metal frame without a great deal of trouble and possible delay.

A series of metal frames with inner containers illustrates the handling and stacking feature (Figure 7) for storing empty containers or for storing material in the containers. This idea may be adapted to eliminate expensive concrete vaults in some cases by merely placing the containers in an isolated room or another area.

You will also note that these metal space frames of slotted steel angle may easily be handled by a fork lift truck or an overhead crane yet are light enough that two men can easily move them a short distance by hand if necessary.

Along with our development effort on metal space frames, we have also investigated different designs for the inner container. The important characteristics that we have considered for the inner container are as follows:

1. Strength to withstand damage from a possible carrier accident while material is in transit.
2. Absolute water tightness.
3. Economy.
4. Ease of access when loading or unloading contents.

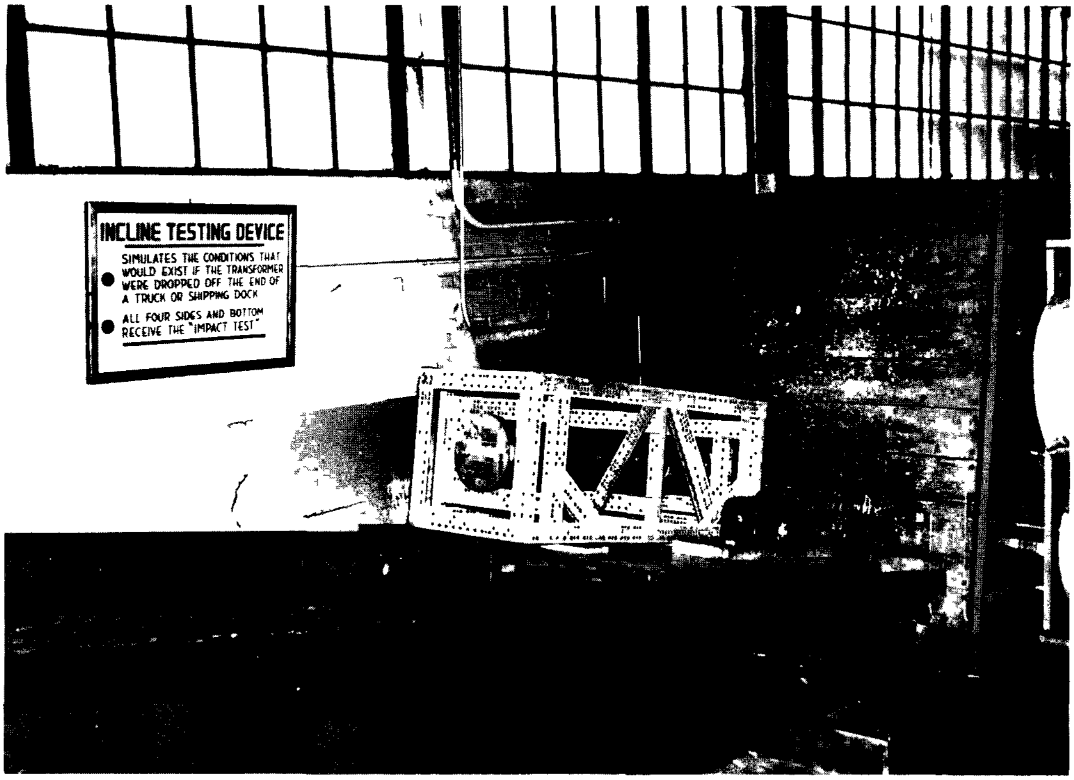

Fig. 5—Incline test results quite satisfactory.

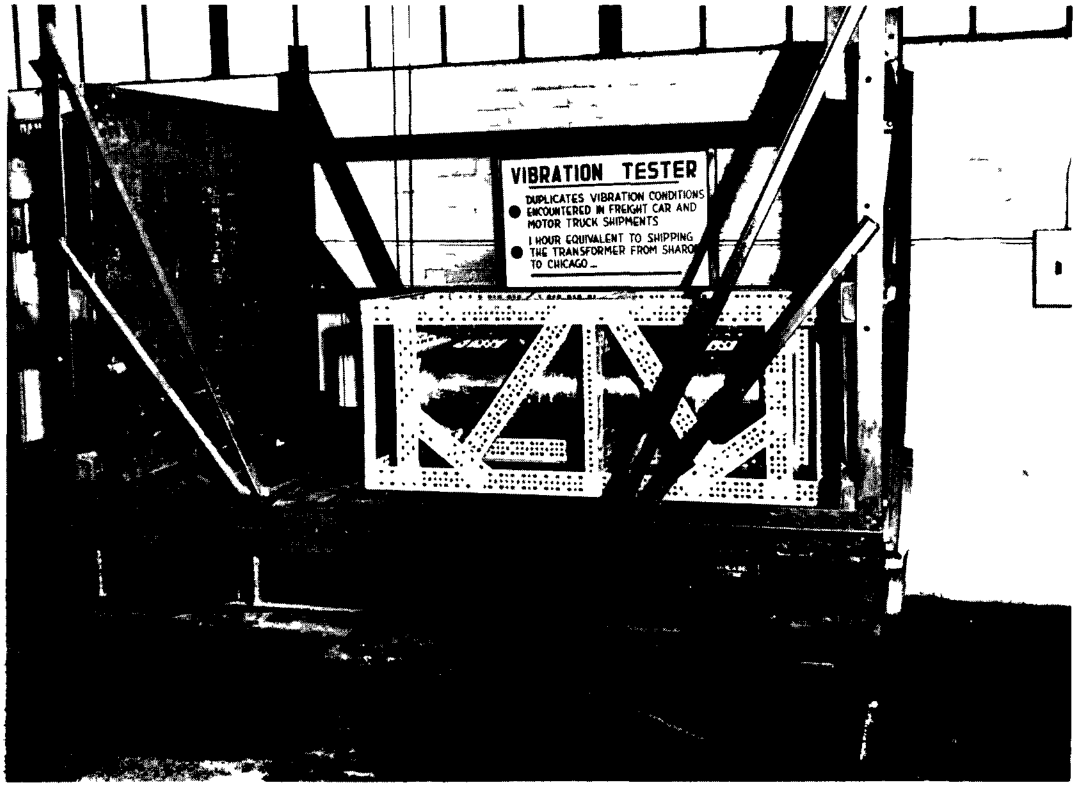

Fig. 6—Vibration test. No loose members noted, results quite satisfactory.

Fig. 7—Storing and handling.

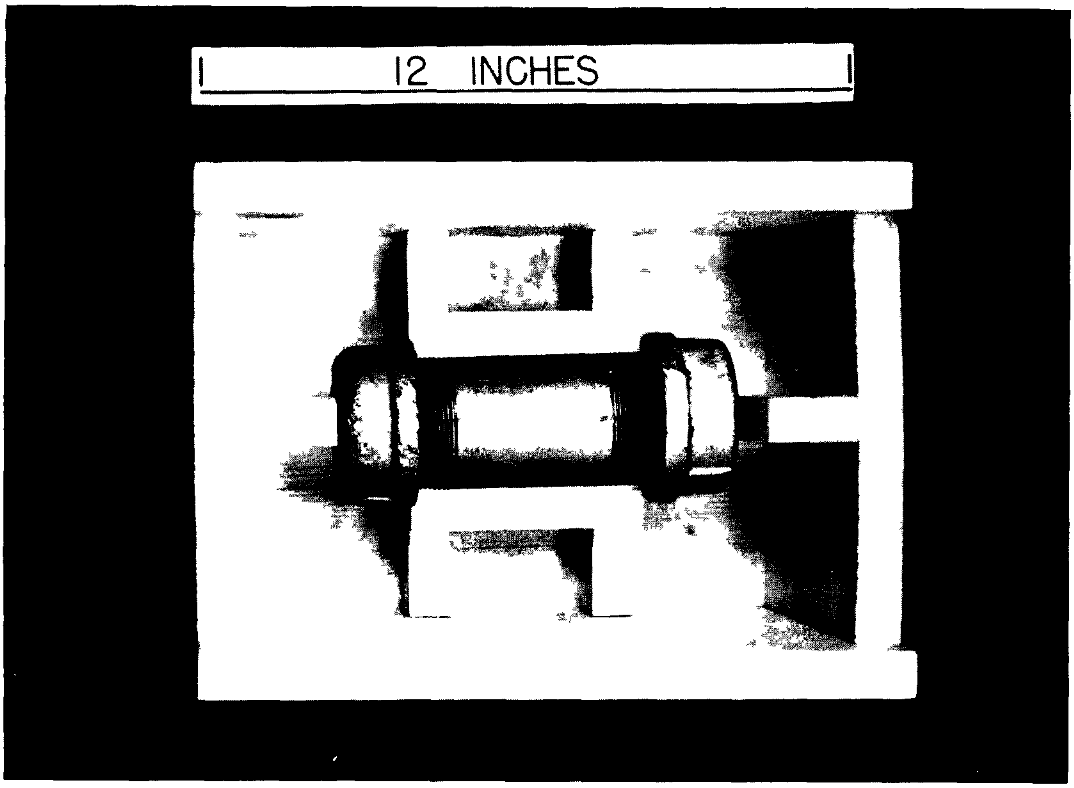

Fig. 8—Three-in long pipe nipple closed off with 2 standard end caps.

5. Availability.

6. Light weight.

The inner container that best satisfied these requirements was then placed in the custom made slotted angle space frame. The result was a light weight, strong, and economical shipping container.

I should like to discuss briefly a few of these other inner containers that we have used.

A. Figure 8 illustrates a 3" long ordinary pipe nipple threaded at both ends. A vial containing a small quantity of enriched material was placed in the pipe nipple and closed off by standard pipe caps. Cost, approximately $2.00.

B. Figure 9 illustrates 3 different types of inner containers.

   1. A standard type container is used by the aircraft industry to package delicate materials for overseas. This unit has limited application to our needs, but is carried in small quantities for possible use. Size, 6-1/2" dia. x 17" deep. Cost, $4.85.

   2. We have also fabricated a container of ordinary pipe with a square steel plate welded to the one end to form the bottom. The top is a square piece of steel plate slotted on each corner with a rubber gasket attached to the under side. The closing device consists of four (4) wing nuts and bolts welded to the side of the container. The bolts swing into the slots of the cover and tighten the cover down completely. Size 6" dia. x 14" high. Cost, $33.00.

   3. An ordinary conduit box used primarily by the electrical industry has recently been adopted. This box is constructed of 14 gage steel. Slight modifications have been made to satisfy our requirements for a strong metal box 4" x 4" x 8" deep. Cost, $10.50.

Each of us have serious obligations to fulfill in controlling SS Material and arranging for transportation within a plant or to a distant destination and providing safe storage. Some of these obligations are as follows:

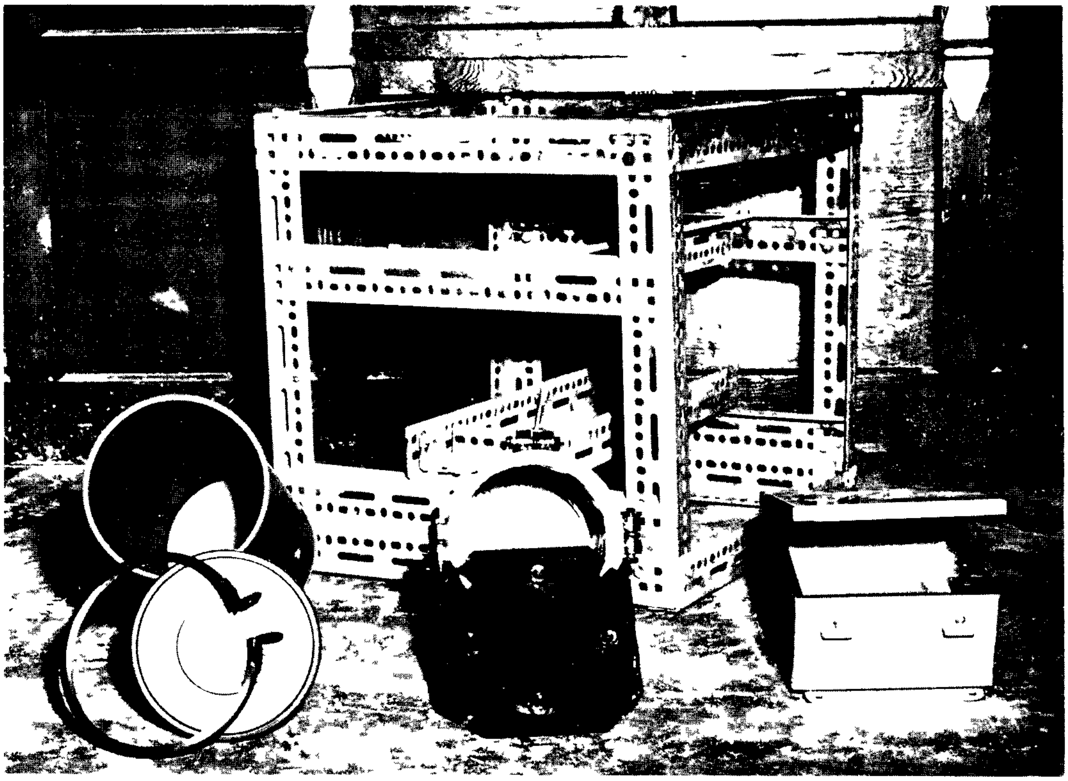

Fig. 9—1. Instrument container used for overseas shipments. 2. Pipe and welded steel plate with lid slotted for closing. 3. Electrical conduit box.

1. We must protect the SS Material that we are responsible for.
2. We must offer for transportation only those shipments that are properly packaged, labeled, and absolutely safe.
3. We must prevent a criticality incident while a shipment is in transit by careful selection of containers that we use. We must construct containers in such a way that shipments made by other companies on the same truck or train will be kept properly separated.
4. We must endeavor to standardize containers whenever possible in order to be consistent in our shipping practices.
5. We must attempt to reduce shipping costs and strive for regulations that satisfy the needs of all.

Conclusion

It is hoped that the information presented herein will stimulate constructive thinking toward producing adequate and standard shipping containers at less cost so that significant contributions may be made to reducing the cost of atomic power without sacrificing safety.