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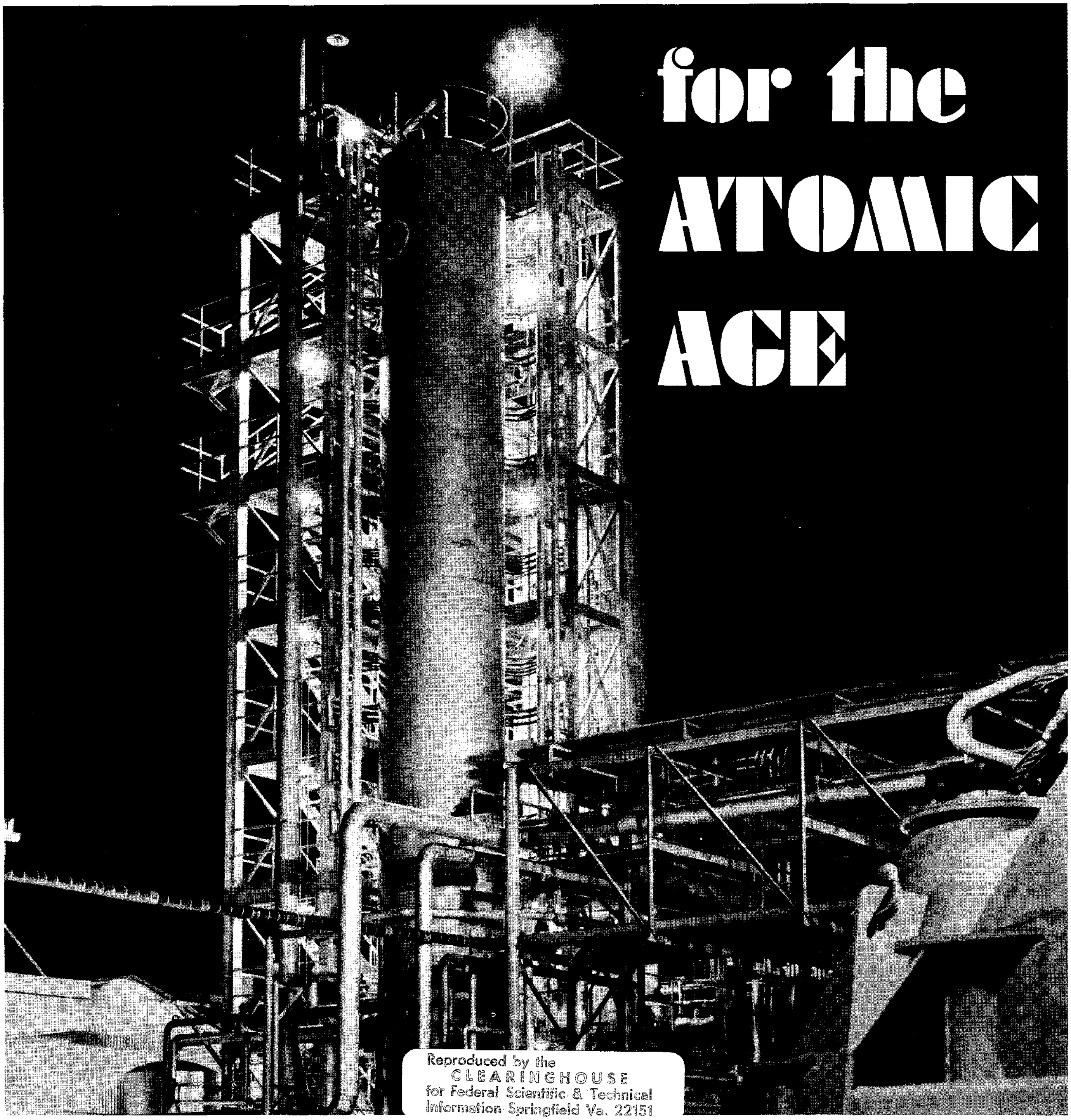
FUEL for the ATOMIC AGE



Completion Report On St. Louis-Area Uranium Processing Operations, 1942-1967

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This report was prepared by Fleishman-Hillard, Inc., St. Louis, Mo., under Mallinckrodt Chemical Works - Uranium Division Subcontract 25188-M of July 11, 1966, as modified. The report was prepared in connection with Mallinckrodt Chemical Works' performance under Contract No. W-14-108-Eng-8 (Principal Contract), as modified, with the United States of America acting through the United States Atomic Energy Commission.

FUEL FOR THE ATOMIC AGE

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Completion Report On St. Louis-Area
Uranium Processing Operations, 1942 - 1967

September 30, 1967

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(General Summary)

Section I

FUEL FOR THE ATOMIC AGE

An Introduction to the History
of Mallinckrodt Chemical Works' Uranium Production
and Development Activities
for the United States Government

"The story of the supply of uranium
is by itself a thrilling one, and
the production of enough pure metallic
uranium to do our task in time was a
technological and industrial miracle."

---- Arthur Holly Compton*

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* Arthur Holly Compton, <u>Atomic Quest</u> (New York: Oxford University Press, 1956), p. 90.

On December 2, 1942, in the early days of World War II, the atomic age was born: the first self-sustaining, nuclear chain reaction was achieved in what had been a squash court under the West Stands of Stagg Field at the University of Chicago. On that day, ". . . man first liberated and controlled the power within the atom. This event was known only to a few. To those few it was a turning point in history, the birth of a new era."*

The historic and dramatic accomplishment on that cold, winter Wednesday in Chicago was the successful result of the combined work, talents, and skills of scientists, engineers, technicians, and others working on related, super-secret projects in various parts of the nation.

One of the most important parts of the hush-hush scientific-industrial complex was the uranium project at the Mallinckrodt Chemical Works plant in St. Louis, Missouri. Its work was a vital link in the chain of activities which led to the birth, and subsequent development and advancement of the atomic age.

This document is a history of the Government's uranium operations in the St. Louis area. Officially, it is a "completion report" describing Mallinckrodt's operations from July 1, 1942, through June 30, 1967, under Contract No. W-14-108-ENG.-8 for the United States of America. To provide perspective, the report discusses significant events and activities from Martin Heinrich Klaproth's discovery of uranium in 1789 to Mallinckrodt's initial involvement in uranium-processing research in the spring of 1942. It continues through the termination of the Company's standby contract with the AEC in June, 1967.

Figure I is a chart summarizing the major contributions of the Mallinckrodt organization to the Government's uranium processing and development efforts in the St. Louis area during the 25-year period from July, 1942, through June, 1967.

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*Ibid., p. 139.

TWENTY-FIVE YEARS OF URANIUM PROCESSING AND DEVELOPMENT

MAJOR CONTRIBUTIONS BY MALLINCKRODT CHEMICAL WORKS

* First Commercial Process for Ether Extraction of Uranyl Nitrate
* First Factory Production of Orange Oxide from Uranyl Nitrate
* First Factory Process for Producing Brown Oxide
* Early Production of Green Salt
* Early Commercial Reduction and Casting of Uranium Metal
* First Stirred-Bed Reactor for Continuous-Process Production of Green Salt and Brown Oxide
* First Commercial Continuous Ether-Extraction Process
* First TBP-Kerosene and TBP-Hexane Processes for Uranium-Ore Refining
* Numerous Advances in Uranium Metal Production, Including Slag Liner, Dingot-Extrusion, and Electrolytic Reduction
* First Successful, Commercial Fluid-Bed Denitration System
* First Integrated, Continuous-Process Fluid-Bed Uranium Production
* Adaption of Uranium Processing Equipment to Commercial Production of Purified Dense Thoria
* Continuous Cost Reduction Through Advances in Manufacturing Practices and Scrap Recovery
* Consistent Fulfillment of AEC Production Objectives

Figure I

Objectives Of The Report

The objectives which guided the preparation of this report were:

(1) To provide a single document describing the major aspects of the technical and administrative history of the Government's uranium processing operations in the St. Louis area.

(2) To explain the rationale underlying the technical and administrative developments of the operations.

(3) To produce an evaluative instrument for comparing and appraising present and future operations of a similar nature.

(4) To provide a general guide for the establishment of similar operations in the future.

(5) To organize a one-source reference on the highlights of the first major processing contract to be phased out in line with the Government's evolving requirements.

(6) To contribute to the literature in the fields of uranium-production technology, nuclear science, chemistry, and technical and administrative management.

The report is written in a semi-technical tone to make it meaningful and useful not only to persons with technical backgrounds, but also to those whose orientations are not primarily technical.

Summary And Organization Of The Report

This report consists of five major sections.

This section, Section I, is an introduction and general summary of the entire document.

Section II, which comprises the bulk of the report, is a history of the philosophy behind the major technological developments of the St. Louis-area uranium operations. The emphasis of the material in Section II is on the rationale underlying the developments rather than on detailed descriptions of the developments themselves. The detailed descriptions can be found in a variety of other technical reports and documents.

Because the uranium effort in the St. Louis area was primarily technical in nature, the history of the operation is, in large part, a technical history.

It began in April, 1942, when Dr. Arthur Holly Compton, Dr. Norman Hilberry, and Dr. Frank H. Spedding approached Edward Mallinckrodt, Jr., to seek his Company's assistance in preparing the extremely pure uranium compounds which were needed as fuel for an experimental atomic reactor at the University of Chicago. The reactor, if successful, would achieve a self-sustaining nuclear chain reaction.

The whole project was of extreme importance to the national security. At the time, the United States had been engaged in World War II for nearly a year, and the nuclear reactor experiment had the potential for making a major contribution to the war effort. A successful nuclear fission reaction, on a proper scale, would release an incredibly enormous amount of energy, and could produce an explosion of immense proportions. The possibility that scientists of the Axis powers might develop a device to achieve such a frightening explosion made imperative a vast effort -- the Manhattan Project -- within the United States to develop such a device first.

It was in this tense, wartime environment that Mallinckrodt was asked to produce the key uranium compounds which were needed before further progress could be made. Dr. Compton and his associates at the University of Chicago already had approached several other major chemical producers to ask their assistance, but they all declined -- partly because of other wartime commitments, and partly because of the difficulty and risk involved in the uranium-purification assignment.

To produce the needed uranium fuel, impure uranium concentrates would have to be purified by extraction with ether. Never before had the extraction been achieved on anything but a laboratory scale, and even on that small scale, the explosive and erratic nature of the ether made the operation extremely hazardous.

Dr. Compton turned to Mallinckrodt because he was familiar with the Company's outstanding reputation for safely producing high-quality, high-purity products, and because he knew that the Company was expert in handling ether.

Mallinckrodt accepted the challenging assignment, and within 50 days, the Company accomplished the "remarkable achievement" of producing highly purified uranium oxide on a tonnage scale.

At that time, the Company's uranium products included, uranium trioxide (UO_3, or orange oxide), and uranium dioxide (UO_2, or brown oxide). Later in 1942, Mallinckrodt started production of uranium tetrafluoride (UF_4, or green salt). When the first self-sustaining nuclear chain reaction was achieved on December 2, all of the uranium in the pile was in the form of uranium dioxide produced by Mallinckrodt or uranium metal prepared by others from intermediate materials produced by Mallinckrodt. A few months later, in July, 1943, Mallinckrodt started its first metal plant.

During the 25 years that it was involved in uranium production, Mallinckrodt made numerous contributions to uranium-processing technology. This report discusses most of the Company's major contributions including the development of the first commercial process for ether extraction of uranyl nitrate; the first continuous ether-extraction process; the tributyl phosphate-hexane process for uranium purification; the pot process for converting uranyl nitrate to orange oxide; the fluid-bed denitration system for producing UO_3; the first factory process for producing brown oxide; improved methods for batch-type, factory production of green salt; the continuous-process, stirred-bed reactor for producing green salt and brown oxide; advances in continuous-process production of green salt by means of the Mallinckrodt integrated fluid-bed system; improved methods for casting and reducing uranium metal; and the dingot and electrolytic reduction processes for producing uranium metal.

Section III is a history of the administrative development of the St. Louis-area uranium operations beginning with the initial efforts in 1942 in Mallinckrodt's building 25-2 research laboratory and continuing through the final activities at the AEC's Weldon Spring facility in 1966.

The subjects discussed in Section III include organization and management, Mallinckrodt's experience and qualifications, contractual arrangements, physical facilities and plant services, material accountability, plant security, and health and safety.

Section IV deals with production and costs. In addition to discussing cost accounting, the section provides an interpretive narrative concerning Mallinckrodt-AEC negotiations, and other pertinent subjects related to production and costs.

Section V is a brief statement summarizing the conclusions of authorities concerning the significance of the work accomplished since 1942 by the Government's St. Louis-area uranium operations.

For almost a quarter of a century, Mallinckrodt successfully carried out its uranium-processing contracts with the Government. The Company, always surpassing its commitments under the requirements of the contracts, continually worked to lower costs, increase production and improve quality.

In April, 1966, when the AEC announced its plans to terminate its St. Louis-area uranium production activities, Dr. Glenn T. Seaborg, Chairman of the Commission, praised Mallinckrodt for its excellent performance in the Government's atomic energy program. He called attention to the Company's outstanding record of accomplishment for production operations and related process improvement and development programs. Dr. Seaborg said that Mallinckrodt "can be justly proud of the important role it has played in the advancement of peaceful uses of atomic energy and in our nation's defense efforts."

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Section II

Technological Developments

Section II
Part 1

COMMERCIAL REDUCTION AND CASTING OF URANIUM METAL

The continuing effort to produce uranium metal has been the single unifying element in both the history of the chemistry of uranium and the history of Mallinckrodt Chemical Works' involvement in uranium production technology.

Therefore, to provide a proper perspective for the entire technical history of uranium production activities in the St. Louis area, it is appropriate to begin this section of the Completion Report by discussing Mallinckrodt's role in uranium metal production -- even though the Company's initial activities did not include production of the metal itself.

Mallinckrodt has played an important part in uranium metal technology in the United States since the first serious efforts were directed towards the development of a commercial production process.

The firm first became involved in the uranium-metal development program in April, 1942, when the Company started work to develop a process for preparing pure uranium dioxide (UO_2, or brown oxide), one of the key intermediate materials required in the preparation of uranium metal.

In July, 1943, the Company started a plant to produce the metal itself. In so doing, Mallinckrodt became one of the first industrial concerns in the nation to produce uranium metal commercially. Since then, the firm's Uranium Division has continued to make important contributions to improving the technology of uranium metal production.

Background

The technology of commercial uranium metal production as practiced by Mallinckrodt -- and by other industrial firms in both the United States and abroad -- was influenced significantly by earlier efforts to prepare uranium metal.

The first successful preparation of uranium metal was achieved in 1841 by Eugene Melchior Peligot, the French chemist. Peligot prepared the metal by the thermite reduction of uranium chloride with potassium metal.

Martin Heinrich Klaproth, who discovered the element uranium in 1789, had first tried to prepare the metal by reducing uranium oxide with carbon; however, Klaproth's experiments did not yield the free metal. More than a century later, in 1893, the French chemist Henri Moissan reported success in obtaining the metal by using the same reaction, but at higher temperatures than those employed by Klaproth.

It was not until the 1920's that research on uranium metal preparation began to arouse the interest of scientists in the United States.

In 1926, scientists at the University of New Hampshire reported the results of research on calcium reduction of uranium oxide and of uranium tetrachloride. In part of the work the researchers employed a bomb-reduction process in which an alundum crucible within a steel bomb was charged with a mixture of uranium tetrachloride and calcium metal. The bomb was heated by electrical resistance heaters to the ignition temperature of the charge, and the reaction fused the uranium metal and calcium chloride products. A massive lump of uranium, which weighed about three pounds, was recovered, and according to the limited analytical data that was published, the metal was apparently of good quality. This process, known as the "James Process," was the first uranium metal process capable of producing massive metal of possibly good quality by simply preheating the charge. The method has large-scale potential; however, it involved several factors that posed major disadvantages.

In the following years, researchers at Westinghouse Electric Corporation and Metal Hydrides, Inc., experimented with other techniques of producing uranium metal. Production was on a very small scale, however, and by 1939, the total amounts of uranium metal produced by both of these companies probably did not total more than 10 pounds.

Between 1939 and 1941, however, the preparation of uranium metal suddenly became significant. Otto Hahn and F. Strassman reported that the results of their experiments on neutron bombardment of uranium were explainable by a fission process. When other researchers in both the United States and Europe checked the experiments, nuclear fission was confirmed, and some scientists envisioned atomic energy being achieved through a self-sustaining, fission chain reaction. Subsequently, a number of scientists, believing that the release of enormous energy tied up in the atom could become a major factor in our national security, urged that research on nuclear energy be strongly supported by the Federal government.

Gradually, more Federal attention was given to the support of an atomic-energy program; however, there was relatively little effort towards the procurement of uranium metal. Uranium metal production in the United States still was measured in quantities of grams or ounces as of December 6, 1941. "At this point," commented H. A. Wilhelm of Ames Laboratory in his report on the history of uranium metal production in America, "just 100 years after Peligot first prepared metallic uranium, the demand for that metal skyrocketed to pound quantities, with earliest possible deliveries."

Initial Mallinckrodt Involvement

Thus, early in 1942 uranium metal operations were expanded, and the first serious attempts were made to develop a practical process for producing uranium metal of nuclear-reactor quality.

In connection with these efforts, Mallinckrodt was asked to attempt to develop a commercial uranium-purification process based on the solubility of uranyl nitrate in diethyl ether. (For additional details concerning Mallinckrodt's early work in this area, see Section II, Part 2.) The Mallinckrodt process initially was to go as far as producing uranium dioxide. Others were called upon to develop methods for reducing the oxide to metal.

Some of the methods for producing the metal required uranium tetrafluoride (UF_4, or green salt) as an intermediate material. To supply this material, green-salt production processes were started at E. I. du Pont de Nemours Company, Inc., and Harshaw Chemical Company late in the summer of 1942. Mallinckrodt also started UF_4 production later that year. (See details in Section II, Part 8.) Green salt was then available from three producers -- all using Mallinckrodt brown oxide.

The first self-sustaining chain-reaction nuclear pile was operated successfully beneath the West Stands of Stagg Field at the University of Chicago on December 2, 1942. All of the uranium used in the pile was in the form of compressed UO_2 produced by Mallinckrodt or uranium metal prepared by others using intermediate, purified uranium compounds produced by Mallinckrodt.

Evolution Of The Basic, Commercial, Metal-Production Process

The Stagg Field pile was the result, in large part, of work by the Plutonium Project, which was organized and directed at the University of Chicago by Arthur Holly Compton with the assistance of Richard L. Doan and Norman Hilberry. The project was given the code name "Metallurgical Project" to provide a wartime disguise for the organization's nuclear development studies. The project laboratory, which was directed by Richard Doan, was named the "Metallurgical Laboratory."

When the Metallurgical Project was being established in February, 1942, Dr. Frank H. Spedding* of Iowa State College, Ames, Iowa, was invited to participate and to direct the chemical research and development activities in Chicago. In addition, a program involving metal-preparation studies was established at Iowa State College. Later, melting and casting of uranium were also studied at Ames.

By mid-June of 1942, the Iowa State group had demonstrated that graphite could be used -- without excessive amounts of carbon pickup -- to contain uranium metal during vacuum-induction melting and casting.

During most of the remainder of 1942, the melting and casting at Ames were done by a process called "drip casting." The metal charge was placed on a graphite grill that was located at the bottom of a graphite crucible which, in turn, was seated over a graphite mold. The crucible-mold unit was induction heated under a vacuum in a large silica tube. The assembly was maintained in a vertical position with the induction coil placed around the outside of the silica tube. During the melting-casting process, the liquid metal flowed from the crucible, through the grill, and into the mold which was heated to a temperature near the melting point of uranium. Oxide film, skulls (thin layers of metal), and oxide sponge were held by the graphite grill. After casting was completed, the assembly was cooled to room temperature and then dismantled to remove the ingot.

Although a satisfactory method of melting and casting had been developed, it was still necessary to develop a practical uranium-metal preparation process that could be applied on an industrial scale.

In the early work on metal preparation at Ames, uranium oxide was used with the carbon reduction method employed earlier by Moissan; however, the carbon contained in the metal caused an undesirable brittleness in the product. Other reductants were tried with the uranium oxide, but none resulted in a satisfactory uranium-metal preparation process.

In the summer of 1942, the group at Ames obtained a small quantity of uranium tetrafluoride from the Metallurgical Project in Chicago. The Iowa State researchers attempted to reduce the material to metal using calcium as the reductant. At about the same time -- August, 1942 -- C. J. Rodden, at the National Bureau of Standards, also conducted experiments on the reduction of UF_4 with calcium. Both the Rodden experiments and the Ames experiments were successful.

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*Dr. Spedding was one of the original group of three -- the others were Dr. Hilberry and Dr. Compton -- who initially contacted Edward Mallinckrodt, Jr., to request his company's participation in the project in April, 1942.

Subsequently, efforts on the green salt-calcium process were expanded at Ames. By mid-September, 1942, with additional green salt available, the Ames group was able to increase the size of the UF_4 charges and, thus, obtain uranium yields averaging better than 90 per cent in massive pieces of metal which were code-named "biscuits."

To complete the process, several biscuits were melted and cast, by the drip-casting method, into a single ingot. Ingots, which were cast as solid cylinders, approximately 2¼ inches in diameter, were cut into sections two to three inches long. The sections were code-named "eggs."

When the chain-reaction pile at the University of Chicago was first operated, it contained nearly equal amounts of metal from Westinghouse, Iowa State College and Metal Hydrides, and several tons of pressed cylinders, also referred to as "eggs," of uranium dioxide produced by Mallinckrodt.

In November, 1942, the Ames group began experiments to examine the possibility of substituting magnesium (Mg) for calcium in the bomb-reduction process. Magnesium, if it could be used, would offer a number of advantages over calcium. These advantages were:

- Commercial-grade magnesium would be less likely to contaminate the metal.
- The weight of the magnesium required for reduction would be approximately three-fifths the weight of the calcium required.
- The cost of the magnesium would be approximately five to ten times less than the cost of calcium.
- The magnesium was much easier to obtain than calcium.

Experimental development on the magnesium process proceeded well, and after January, 1943, the entire metal-production operation at Ames employed the magnesium reduction technique. Another change made at Ames was in the casting operation. The original drip-casting process for remelting and casting was replaced with a process using larger equipment, and a crucible containing a valve for pouring the liquid metal into the mold.

Three industrial firms, including Mallinckrodt, studied the metal-production processes developed at Ames and used them as guides in planning their own metal-production operations.

Industrial-Scale Metal Production Process At Mallinckrodt

Mallinckrodt started its first metal plant in July, 1943. The bomb-reduction operations initially employed by Mallinckrodt were adapted from the basic uranium tetrafluoride-magnesium bomb-reduction process used at Ames. There were, however, significant variations from the Ames process in the remelting and casting operations.

The first major step in the metal production process at Mallinckrodt was preparing the reactor vessel -- a steel bomb shell -- for thermite reduction. Preparation of the bomb began by lining it with a refractory material. The material used was electrically fused, low-boron dolomitic oxide ($CaO.MgO$). After the bottom of the bomb had been covered with the refractory, a tapered steel mandrel, with its diameter increasing from bottom to top, was positioned in the center of the bomb shell, and the annular space between the mandrel and the shell was filled with liner material. Pneumatic jolting was used to pack the refractory tight against the bomb-shell wall.

The next steps in the Mallinckrodt metal-preparation process were charging and closing the bomb. First the steel mandrel used in lining the bomb was cautiously removed from the reactor shell. Then the charge, containing uranium tetrafluoride intimately blended with magnesium chips, was fed into the lined cavity and tamped. Next, a suitable thickness of the same refractory material used to line the side of the bomb was packed on top of the UF_4-Mg charge. The capping operation was completed by bolting a solid-steel cover plate onto the top of the bomb shell.

After capping, the bomb was placed into a top-loading furnace for firing.

Although the reaction between uranium tetrafluoride and magnesium is exothermic, the heat of the reaction is not sufficient, by itself, to provide the fusion necessary to achieve adequate separation of the reaction products -- uranium metal and magnesium fluoride. Therefore, in the UF_4-Mg reduction process, the entire bomb was pre-heated in a furnace to provide the additional heat required to achieve complete fusion and separation.

> (Under these circumstances, thermite reduction occurs when the temperature of the bomb reaches approximately the melting point of the magnesium. While the products of the reaction are in their molten states, the dense liquid uranium metal sinks to the bottom part of the bomb and collects in a pool, and the magnesium-fluoride by-product -- the slag -- collects on top of the pool of liquid metal.)

After this reaction had taken place, the bomb was removed from the furnace and cooled to room temperature.

The cooled bomb shell was then transferred to an unloading or "break-out" area where the shell was opened and the products within were removed. A pneumatically operated jolter was used to jolt the shell and, thus, discharge the entire contents. The solidified metal was removed as one large massive regulus, which was referred to as a "derby," a code name replacing the previous code identification which was "biscuit."

Magnesium slag adhering to the metal was broken or chipped loose, and the cleaned "derby" was then weighed. For accountability records, the yield from the reduction operation was determined by the weight of the chipped derby. The reaction vessel was cleaned and returned to the loading line where it was prepared for another "run." The life of a bomb shell was approximately 150 runs.

Melting And Casting Innovations

Up to this point, the pattern of the metal preparation process at Mallinckrodt closely followed that of the operations at Iowa State College. Mallinckrodt's melting and casting operations, however, varied considerably from the Ames melting and casting process which initially employed drip-casting equipment and was later modified for pouring.

The melting-casting process at Mallinckrodt was carried out in an induction-heated, large vacuum vessel. The induction coil, which was positioned in the center of the vessel, surrounded a graphite melting crucible and was thermally and electrically insulated from it. A pouring hole, which was kept closed mechanically during melting, was incorporated into the bottom of the melting crucible. Graphite molds were mounted in the vacuum tub below the bottom of the melting crucible.

In the melting process, the charge metal in the crucible was melted by induction heating under high vacuum. After the charge had melted, the pour hole was opened mechanically, and the liquid metal poured into the graphite molds.

Three Major Process Improvements

Following construction in 1950 of a modernized and mechanized metal plant, Plant 6E at Destrehan Street, development work at Mallinckrodt resulted in three major changes in the metal processing operations. These changes -- which were made to increase production, improve quality, and reduce costs -- involved: (1) increasing the size of derbies and cast ingots; (2) carrying out the casting operation to encourage vaporization of impurities from the remelt crucible and to promote "hot-topping" of the ingot mold; and (3) using recycle slag as refractory liner instead of dolomitic lime.

Larger Derbies And Ingots

During 1953, development work at Mallinckrodt's metal pilot plant revealed: (1) that it would be feasible to increase the amount of UF_4 charged to a bomb shell for reduction to metal; and (2) that it would be feasible to increase the size of the metal charge to the casting furnace.

While these pilot-plant studies were still in progress, the production plant undertook an investigation to determine how it could increase its capacity by 50 per cent. After careful study it was concluded that the increase could be achieved by modifying equipment so that it could operate with larger unit charges in both the reduction and casting steps.

To obtain the extra capacity, the reduction furnaces had to be modified to produce approximately 300 pounds of metal per charge instead of the 120 pounds previously produced. This change increased the size of each derby from about eight inches in diameter by four inches in length to about 12 inches in diameter by four inches in length. The charge for the casting furnace was increased from 640 pounds to about 1225 pounds. This change increased the size of the crude ingot from five inches in diameter by 45 inches in length to seven inches in diameter by 45 inches in length.

Vaporization Of Impurities And "Hot-Topping" During Casting

The results of an extensive chemical and physical investigation by Mallinckrodt of the distribution of impurities in remelted uranium metal led to the development of a major process improvement in the melting and casting of uranium metal.

It was found that the quality of ingots could be improved by reducing the impurities in the metal through vaporization and reducing both the impurities and the shrinkage voids by "hot-topping."

Impurities from the remelt uranium were vaporized during the metal casting process by superheating the molten metal in the remelt crucible several hundred degrees. The melting point of uranium of 99.99 per cent purity is 2071.4°F. By vacuum pumping and heating the furnace to 2650°F, and allowing the melt to outgas at this temperature for 45 minutes, it was possible to distill residual magnesium, some slag, hydrogen, radioactive decay products, and other volatile impurities from the melt.

"Hot-topping" involved controlling the casting operation to cause the ingot to solidify from bottom to top, and thus to cause impurities in the poured metal to float to the top of the mold. This was accomplished by creating a steep temperature gradient in the mold.

In the casting equipment, the ingot mold was positioned directly under and in very close proximity to -- almost touching -- the remelt crucible. The upper part of the mold was insulated, but the bottom portion was not. When the metal charge was melted, the heating, together with the insulating sleeve around the upper part of the mold, created a steep temperature gradient from the top to the bottom of the mold. When the melt was poured into the mold, the temperature gradient caused solidification to proceed from the bottom to the top of the ingot.

This casting practice normally produced ingots with practically no internal cavities, and the directional solidification caused the nonmetallic impurities to concentrate at the top of the ingot in a shallow layer that subsequently could be cropped.

The Use Of Recycle Magnesium Slag As Bomb Refractory Liner

From 1942 until 1954, Mallinckrodt used electrically fused, low-boron dolomitic oxide as refractory liner for the bomb shells. Although the dolomitic oxide had good refractory characteristics, it was relatively expensive (costing about $150 a ton), and because it is hygroscopic, it required close control of sampling and moisture content. Excessive moisture in the liner would cause undesirable side reactions -- such as hydrolysis of the green salt to brown oxide -- and, therefore, lower bomb yields.

Seeking a more practical refractory liner, Mallinckrodt began development work in 1952 on the use of the bomb-reduction by-product magnesium fluoride (MgF_2) to line the reduction vessel. The advantages of magnesium-fluoride slag were:

- Fused magnesium fluoride produced by the thermite reaction of UF_4 and Mg is practically nonhygroscopic.

- The cost of the magnesium fluoride refractory involved only the expense of converting the slag to a form suitable for liner. This amounted to about $10 a ton.

- By using the slag as liner, it was possible to avoid contamination of the metal with outside sources of undesirable impurities such as boron.

The company's work on employing recycle slag as bomb liner included the development of a method for crushing and grinding the magnesium fluoride to convert it to a usable form. The development work was successful, and by 1954, recycle Mg slag had permanently replaced dolomite oxide as refractory liner in the bomb-reduction process.

Conclusions

Mallinckrodt has played an important role in the field of uranium metal preparation since the spring of 1942, when the Metallurgical Project at the University of Chicago first requested the Company to prepare certain purified uranium products for use in the chain-reaction pile at Stagg Field.

The Company's first technical contribution was the almost immediate development of a successful commercial purification process to produce the brown oxide required in the production of uranium metal. Shortly thereafter, the Company also installed a process for producing green salt.

In July 1943, Mallinckrodt's role was expanded when, in addition to producing important intermediate materials, the firm started operations to produce the metal itself. Following the basic process developed at Iowa State College, Mallinckrodt became one of the first commercial enterprises in the country to produce uranium metal on an industrial scale. In subsequent years, the firm made ongoing process improvements to increase production, improve quality, and lower costs. Shortly after World War II, Mallinckrodt became the only plant in the United States to continue to produce virgin uranium metal.

Two major innovations in metal production technology developed by Mallinckrodt are covered in separate subsections of this report. They are the "dingot" (direct ingot) metal process, which is discussed in Section II, Part 11, and the electrolytic reduction process, which is described in Section II, Part 12.

Production And Costs

Production and cost details are contained in Section IV.

Related Documents

The following numbers refer to bibliography entries which are the principal sources for more detailed information on the uranium-metal development work described in this subsection: 6, 8, 9, 10, 11, 12, 13, 15, 19, 21, 22, 41, 44, 45, 57, 63, 67, 70, 88, 89, 91, 92, 98, 102, 105, 106, 107, 108, 109, 119, 128, 129, 133, 135, 136, 137, 139, 140, 141, 142, 143, 144, 145, 147, 150, 126, 161, 167, 168, 186, 190, 198, 199, 201, 202, 211, 216, 222, 223, 224.

Additional references may be found in some of the sources indicated above.

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Section II
Part 2

DEVELOPMENT OF THE FIRST COMMERCIAL PROCESS FOR ETHER EXTRACTION OF URANYL NITRATE

The Manhattan (atomic-bomb) Project of World War II provided the stimulus for the first attempts in history to produce uranium materials on a large, commercial scale. Mallinckrodt Chemical Works did the initial uranium-production work, the most important aspect of which was the development of a practical, commercial ether extraction process for purifying uranium.

Background

In the spring of 1942, Dr. Arthur Holly Compton and his colleagues from the Metallurgical Laboratory of the University of Chicago were working on a secret project, part of the now-famous Manhattan Project, to demonstrate a self-sustaining chain reaction in an atomic reactor. Their project required the construction of a "uranium pile."

For the atomic reactor to work, uranium of a very high degree of purity was required. Impurities would act as poisons in the uranium pile: they would inhibit the desired chain reaction by absorbing neutrons. The success of the pile, therefore, depended on having uranium materials which were free of neutron absorbers, especially elements such as boron, cadmium, and the rare earths.

Under these circumstances, on April 17, 1942, Dr. Compton asked Edward Mallinckrodt, Jr., whether he would have his company attempt to prepare the pure uranium compounds urgently needed to complete this project which was so vital to the war effort.

Dr. Compton and his colleagues discussed with Mr. Mallinckrodt the possibility of using ether extraction, a method of purification which had been demonstrated to be adequate on a laboratory scale, although it had never been accomplished on a large, commercial scale.

The difficulty of the task, and the dangers associated with handling ether had led several major chemical producers previously approached by Dr. Compton to decline his request. Mr. Mallinckrodt, however, agreed to try to solve the problem.

The basic laboratory principle had been discovered 100 years earlier by the French scientist Eugene M. Peligot. The immediate task at Mallinckrodt was to attempt to apply that principle on a large scale to purify, in tonnage quantities, the uranium compounds needed.

The Process

Peligot employed the solubility of uranyl nitrate in diethyl ether to obtain uranium that was free of other metallic elements.

The basic process involves dissolving crystals of uranyl nitrate hexahydrate ($UO_2(NO_3)_2 \cdot 6H_2O$, or UNH) in diethyl ether, separating the ether and water layers that form, and evaporating the ether to recover the purified uranyl nitrate.

The purified uranyl nitrate thus obtained can be used as feed for producing uranium oxides which, in turn, can be used to produce uranium metal.

Proven, Experienced, Scientific And Technical Capability Required

After Peligot's initial discovery, a number of investigators had reported the explosive reaction of nitrate with ether, and consequently, the process was considered by many to be dangerous not only on a large scale, but even on a laboratory scale.

Therefore, whoever was to attempt to develop the large-scale ether extraction process to produce the highly purified uranium materials needed by the Chicago group would have to be able to cope with the dangers involved.

Mallinckrodt Chemical Works was asked to attempt the assignment for two chief reasons:

(1) The Company had a reputation for exceptional skill in producing high quality, highly purified chemicals.

(2) Through previous work, the Company was also well-known for its experience and skill in handling ether.

The Initial Mallinckrodt Ether Extraction Process

The formal research project, at first called "Uranium Oxide S.L. 42-17," was officially started at Mallinckrodt on April 24, 1942, only one week after Dr. Compton had initially asked the Company to undertake the assignment. Mr. Mallinckrodt had made an oral agreement with Dr. Compton to work on the job for an initial fee of $15,000. The project name, including the abbreviation "S.L.," was purposely chosen to imply that the uranium compound was simply another of Mallinckrodt's line of standard luminescent chemicals. However, using the term "uranium" was against government security regulations, and the name of the project was changed to "Tube Alloy Dioxide."

Mallinckrodt's primary research and development task was to try to translate the laboratory concept into a large-scale process.

In the process that Mallinckrodt subsequently was successful in developing, the starting material was commercially available black oxide (U_3O_8). The crude black oxide was added to hot concentrated nitric acid in large stainless-steel tanks to produce a crude uranyl nitrate solution. The uranyl nitrate solution was then filtered through a stainless steel filter press and subsequently concentrated in tanks heated by steam coils to the boiling point (248°F) of pure uranyl nitrate hexahydrate. After the molten crude uranyl nitrate was cooled to 176°F, it was pumped directly into cold ether. The water layer and ether layer separated, and the ether layer was washed with small amounts of distilled water. Then the purified material was re-extracted from the ether with relatively large quantities of distilled water. The solution of pure uranyl nitrate, which contained some ether, was heated to the boiling point to drive off the ether.

The subsequent steps in producing oxides were thermal denitration of the uranyl nitrate to produce orange oxide (UO_3), and UO_3 reduction to brown oxide (UO_2).

The Pilot Plant

The urgency of the project required the Mallinckrodt engineers and scientists to utilize existing facilities and available supplies and equipment to develop the process, carry out experiments, assemble plant equipment, and construct the plant in an extremely short period of time.

The pilot plant for the ether extraction development work was an alley between two buildings at Mallinckrodt's facilities in downtown St. Louis. It was there that Mallinckrodt personnel set up an apparatus which, if it worked, was to be the prototype for the large-scale extractor.

The prototype was fabricated entirely of stainless steel. It consisted of a section of pipe, 8 feet long and 2 inches in diameter, to simulate the extractor, and a 3/4 inch pump which mixed and circulated the water and ether. In addition, necessary piping and valving arrangements were provided for the unit which had a capacity of about 1.6 gallons.

The prototype, employing the ether extraction process described above, was successful. It achieved the same level of purification as was achieved earlier on a smaller scale in the laboratory. On May 9, 1942, samples of the uranyl nitrate produced in the prototype extractor were shipped for testing to Princeton University, the University of Chicago, and the National Bureau of Standards. The product passed all existing tests that could be applied for purity.

The Plant-Scale System

The next step was for Mallinckrodt to attempt to design and build a full-size, plant-scale extractor based on the successful prototype of the "pilot plant in the alley."

Again, the project required the Mallinckrodt personnel assigned to the project to use available materials and a variety of design and construction short cuts. For example, for some equipment, rough sketches instead of finished drawings were used together with around-the-clock supervision by the Mallinckrodt engineers. In some cases sketches were not even used. Instead, construction was based on diagrams chalk-drawn on the plant floors or walls.

Fortunately, when the construction was ready to proceed, Mallinckrodt had two buildings available, and they were used to house the plant-scale system. One of the buildings, Building 51, is shown in the inset in Figure II-2.1, which is an aerial view of the main St. Louis plant.

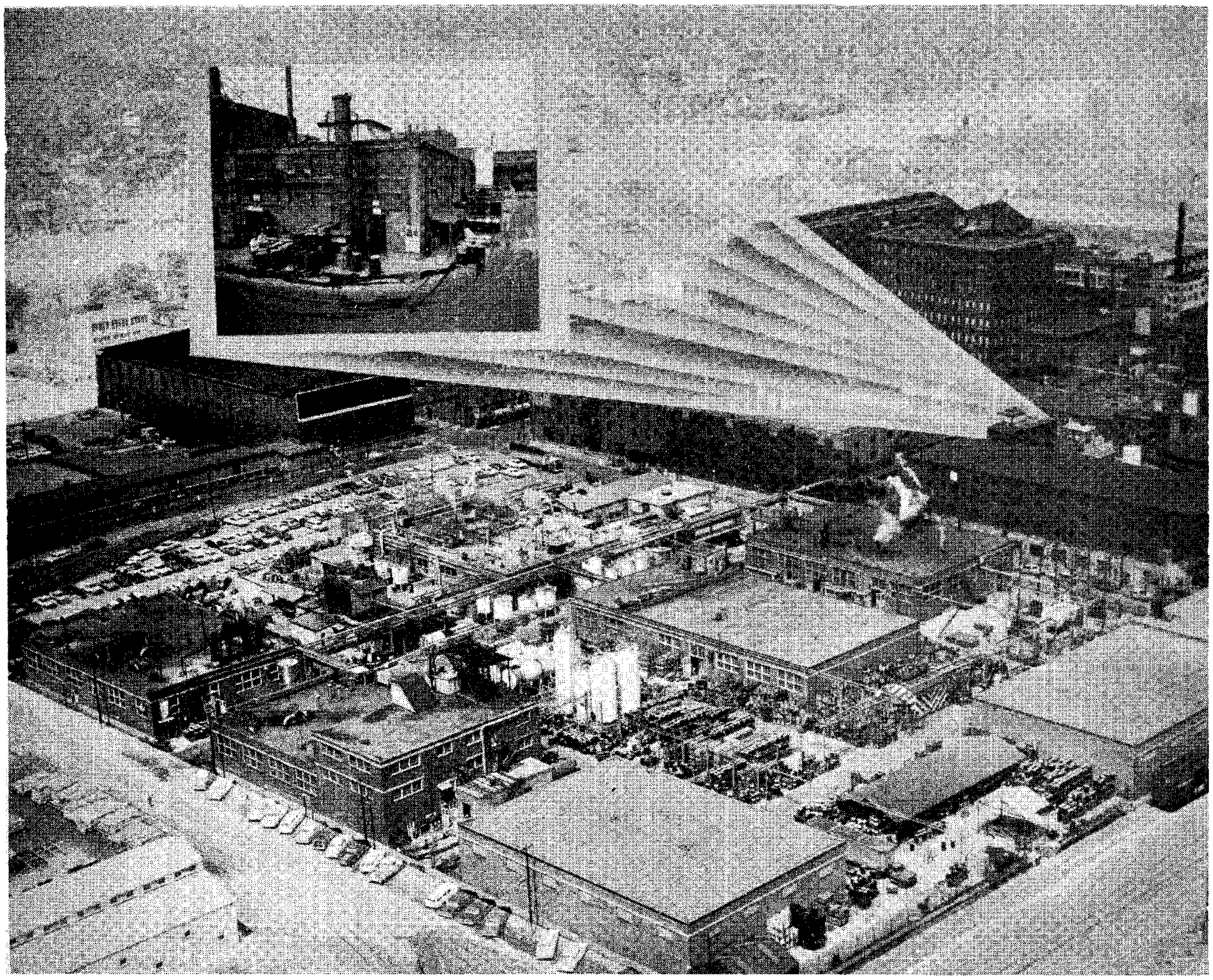

Figure II-2.1 THE LOCATION OF BUILDING 51, one of the two buildings that housed the first plant-scale extractor, is shown in the inset in this aerial view of Mallinckrodt's main plant.

Most of the materials and equipment used in the plant were obtained from a supply which Mallinckrodt had accumulated prior to 1942 in anticipation of possible war-time shortages. The Company had several 1000-gallon stainless-steel tanks and an adequate supply of stainless-steel pipe, valves, and fittings. Mallinckrodt used its own facilities to fabricate most of the other equipment needed.

Start-up and shake-down problems were minimal. Once the processing began, the plant was operated 24 hours a day.

Advantages And Disadvantages For Long-Term Production

Although the initial ether extraction process described here was designed for a short-term project, it was actually used for a four-year period. In evaluating the process from a long-term point of view, several advantages are evident. The plant-scale process required relatively small capital investment, and as a batch operation it also permitted multiple use of equipment such as tanks, pumps, and pipes. Furthermore, it was a simple process and because it required very little instrumentation, it provided additional economies.

Compared with a large-scale continuous process, however, the early ether extraction process had the disadvantages of higher operating costs and lower uranium recoveries. Its main disadvantage was that it did not provide sufficient decontamination of the uranium, and prior processing of the starting materials to relatively pure concentrates was required. The continuous process, described in Section II, Part 3, was developed to overcome this problem.

Conclusions

Mallinckrodt's achievements during the first stage of what was to become a 25-year uranium production project are considered outstanding by authorities associated with the work.

The Company's chief accomplishments during that period were:

(1) Performing the extensive research and gathering the large volume of data necessary to translate a concept, demonstrated previously only on a small laboratory scale, into a workable, plant-scale, tonnage-production process.

(2) Under the handicaps of limited information, time pressures, and wartime shortages, designing a prototype extractor and conducting research, analysis and experiments to prove the effectiveness of the newly developed extraction process.

(3) Under the same handicaps, and with limited sources of supplies and equipment, designing, constructing, and operating a plant for safe, commercial, large-scale ether extraction of uranyl nitrate.

The difficulties of accomplishing these tasks included not only dealing with a number of previously unexplored aspects of nuclear physics, but also the effective and practical meeting-of-the-minds of two different groups -- the University of Chicago physicists, and the Mallinckrodt chemists -- from two distinct scientific disciplines.

The exceptional cooperation between these two groups was demonstrated by the fact that only fifty days from the start of the project, the necessary data were gathered, the plant equipment designed and assembled, the plant built, and the product produced at a rate of more than one ton per day to serve as the sole source of purified uranium for the Manhattan Project well into 1943.

Henry DeWolf Smyth, in his official review of atomic energy work during the war, summed up Mallinckrodt's accomplishments during those first fifty days. He said, "It was a remarkable achievement to have developed and put into production on a scale of the order of one ton per day a process for transforming grossly impure commercial oxide to oxide of a degree of purity seldom achieved even on a laboratory scale."

Production And Costs

Production and cost details are contained in Section IV.

Related Documents

The following numbers refer to bibliography entries which are the principal sources for more detailed information on the development of the first commercial ether-extraction process described in this subsection: 5, 6, 8, 9, 10, 11, 12, 15, 41, 44, 57, 67, 70, 84, 87, 93, 172, 186, 190, 192, 198, 199, 201, 211, 222, 223, 224, 225.

Additional references may be found in some of the sources indicated above.

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Section II
Part 3

DEVELOPMENT OF THE CONTINUOUS ETHER-EXTRACTION PROCESS

In 1945-46, Mallinckrodt Chemical Works developed a continuous ether-extraction process for purifying uranium, and designed and constructed a large, tonnage-scale plant using the process. With continuing improvements, the process remained the standard one at Mallinckrodt's St. Louis operation until 1957 when tributyl-phosphate-hexane was introduced to replace ether.

Background

The original batch ether-extraction process, developed on a crash basis, had several disadvantages. (The batch method is discussed in detail in Section II, Part 2.) Chief among the disadvantages were high operating costs and low percentages of uranium recoveries. Further, removal of certain contaminants from the uranium was insufficient, and the method required relatively pure concentrates, which made extensive prior processing of the starting materials necessary.

The New York Operations Office of the Manhattan District initiated activity to explore the possibility of overcoming these disadvantages. The office assigned to Yale University the task of investigating the possiblities of a continuous ether-extraction technique that would use feed solutions prepared directly from pitchblende by nitric acid digestion.

The Yale work indicated that the approach might be feasible, if a number of complicated inherent problems could be solved.

Initial Development Work At Mallinckrodt

In the spring of 1945, as the Yale studies were being completed, Mallinckrodt began research to develop a final, continuous, counter-current ether-extraction process. Work was aimed at finding a method which would use feed solutions prepared directly from pitchblende ores, or crude concentrates containing 30-80 per cent black oxide.

Mallinckrodt had to solve several complex problems, some of which were revealed in the work at Yale. The major hurdles were to:

- develop satisfactory methods to prepare suitable feed solutions.
- cut down on impurity (especially molybdenum) carry-through during extraction.
- get more complete recovery of uranium.

The total job was two-fold: (1) development of workable methods for preparing suitable feed solutions; and (2) research, development, and design of the continuous extraction process and equipment.

Preparation Of Suitable Feed Solutions

Mallinckrodt worked on developing two methods for preparing feed solutions -- one with pitchblende (containing radium) as the starting material, and one using radium-free concentrates.

The method that Mallinckrodt developed for processing pitchblende involved several steps. First, the original material was ground, and then it was digested with nitric acid. The resulting solution, however, contained radium and sulfate, both of which presented additional problems.

The radium posed a problem both because of its inherent radioactivity hazards, and because of its high value. The sulfate -- if not removed from the solution -- would cause precipitation and mechanical difficulties during extraction.

Mallinckrodt found techniques to overcome these problems. The excess sulfate in the digestion batch was removed by precipitating the sulfate with barium carbonate which also coprecipitated the radium as sulfate. The precipitates were removed by centrifuge techniques, leaving a feed solution with little barium or sulfate.

Mallinckrodt's preparation of feed solution using chemical concentrates free of radium or sulfides and containing only relatively small amounts of sulfate or calcium, involved simply the digestion of the concentrates with nitric acid. The resulting slurry, after adjustment of free nitric acid and uranium content, was used directly as the extraction feed.

One obstacle remained in the preparation of the extraction feed. It was incomplete uranium dissolution. To dissolve the uranium completely required an excess of nitric acid. During the extraction process, the ether stripped the aqueous feed of acid and this caused precipitation to occur. The precipitation, in turn, caused sludging and emulsion which interfered with proper operation of the extraction equipment. Mallinckrodt's solution to the problem was to add a controlled concentration of nitric acid to the ether to prevent the stripping.

Pilot Plant Development Of The Continuous Ether-Extraction Process

In addition to solving the problems associated with developing feed-preparation processes, Mallinckrodt also had to solve major problems in developing the extraction process itself.

In the spring of 1945, a pilot plant was built as an annex to the batch ether-extraction plant then in use. The pilot plant was used to carry out the extensive experimental work leading to development of a practical, continuous, counter-current, dual-cycle ether-extraction process -- one that could be translated into a plant system.

In terms of the physical chemistry involved, the continuous process that Mallinckrodt developed was basically identical to the batch process. An aqueous uranyl-nitrate feed solution is dissolved in diethyl ether. Then the ether and water layers that form are separated, and the purified uranyl nitrate is recovered by further treatment of the ether solution.

The continuous process developed by Mallinckrodt had two cycles: (1) acid extraction, and (2) neutral extraction. The process is illustrated in Figure II-3.1

Acid Extraction

The first stage of the acid cycle was the extraction column, a tube packed with small cylindrical sections of ceramic pipe to promote intimate mixing of the aqueous feed and the ether phases. The ether was the continuous phase and was introduced into the column at the bottom. It flowed counter-current to the aqueous feed which moved by gravity from the top of the column. After passing through the extracting column, the uranium-enriched ether phase overflowed from the top and was carried by pipe to the bottom of a wash column.

The ether solution carried with it traces of impurities, some of which were dissolved, and some of which were mechanically entrained. (Part of the molybdenum, because it is soluble in ether, was one of the impurities that was carried through.)

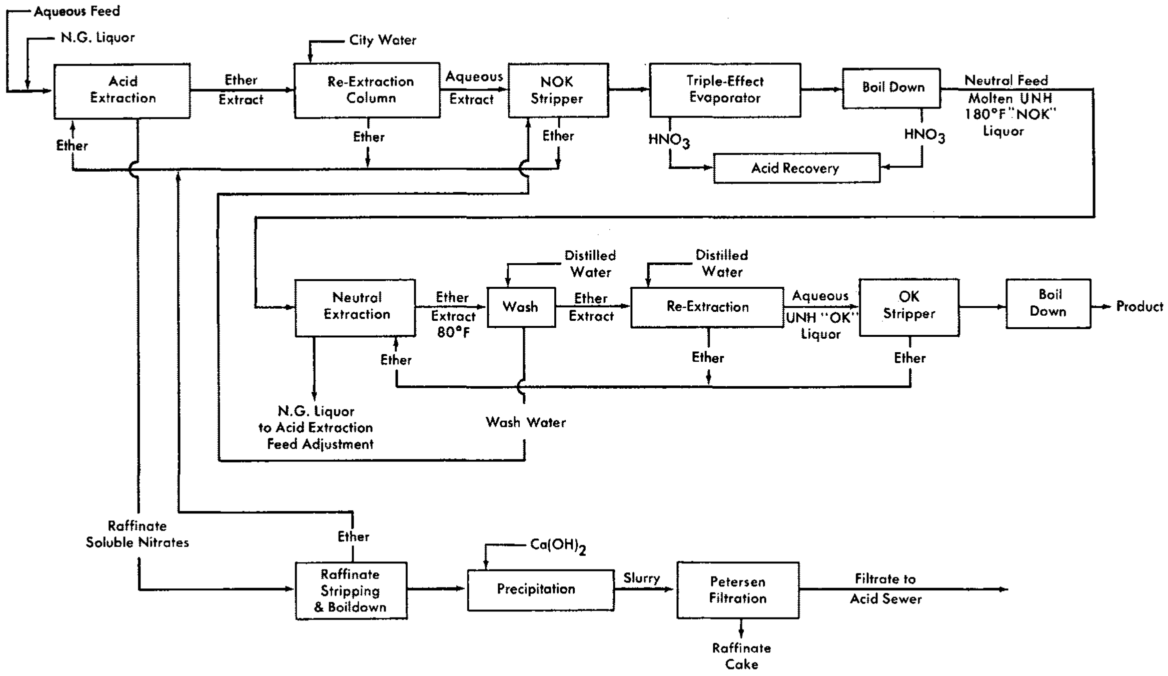

Figure II-3.1 THE FIRST COMMERCIAL CONTINUOUS ETHER-EXTRACTION SYSTEM was developed by Mallinckrodt as a dual-cycle process.

After it left the extraction column, the uranium-rich solvent stream was then passed through the wash column where small amounts of water were used to remove some of the impurities in the solution.

Re-extraction was the final step in the acid-extraction cycle. Large amounts of water were introduced into the top of the re-extraction column. The aqueous phase, flowing by gravity from the top of the column, mixed with the uranium-rich solvent which flowed counter-current to the water from the bottom of the column. As this occurred, the uranyl nitrate, molybdenum, and other remaining impurities were re-extracted into the water phase.

To overcome the problem of impurity carry-through, Mallinckrodt had originally developed a method using charcoal to absorb the molybdenum and phosphorous compounds present in the ether-uranyl-nitrate solution. However, the charcoal method was somewhat cumbersome and expensive. Ultimately, the problem of impurity carry-through was overcome by development of the neutral extraction cycle.

Neutral Extraction

Mallinckrodt developed the neutral extraction process to complete purification of the product produced during the acid extraction cycle.

The first step in the neutral cycle was to boil the uranium-rich water solution (with impurities) that had been produced in the re-extraction column of the acid cycle. The boiling first drove off the small amount of ether remaining in the solution, and then was continued to drive off excess water and acid.

In the process, the heat (280°F-300°F) converted the molybdenum and phosphorous present to compounds which are insoluble in neutral ether. Consequently, during further treatment with ether, the molybdenum and phosphorous impurities would not accompany the uranium.

In the next step of the neutral cycle, the concentrated uranyl-nitrate hexahydrate solution produced by the boiling operation was added to neutral ether on a continuous basis at a high temperature in a mixing chamber. This resulted in the extraction of approximately 85 per cent of the uranyl nitrate into the ether solvent. Fifteen per cent remained with the water phase which was returned to the acid-extraction cycle for recovery of the remaining uranium.

The extract was then washed with a small amount of water to remove remaining impurities. In a final re-extraction column, the uranyl nitrate was extracted from the ether solvent into an aqueous solution. The small amount of ether contained in the water solution was then boiled off leaving purified uranyl nitrate as the final product.

Development Of The Plant-Scale System

Mathematical scale-up based on the pilot plant determined the design of the plant system.

The plant, which was put in operation in 1946, worked very well, and the first product was of the required purity. Aside from a few minor mechanical problems, which were quickly solved, no major difficulties of a technical nature were encountered.

However, one equipment failure resulting from the accidental mixture of strong nitric acid with ether did occur during start-up. The mixture reacted violently, rupturing a tank. Redesign of the part of the process involved prevented recurrences.

Conclusions

In a period of approximately one year, Mallinckrodt carried out extensive laboratory and pilot-plant research and development activities that ultimately resulted in establishing, for the first time anywhere, a continuous, dual-cycle, counter-current ether-extraction process for purifying uranium from crude ore or concentrates. Mallinckrodt effectively translated the process into a successful plant system for producing purified product on multi-tonnage scale.

The continuous process developed by Mallinckrodt had many advantages over the batch process which it replaced. The continuous process required less material handling, less equipment time, and less chemicals for specific rates of production; all of which resulted in lower costs.

In addition, the multi-stage acid extraction resulted in recoveries of more than 99.8 per cent compared with 95 per cent using the batch process. Furthermore, because the process included a second extraction step, the final product was somewhat purer than that produced by the batch plant. Flexibility in the use of starting materials was another distinct advantage of the continuous process.

The chief disadvantages were that, though greatly improved, recoveries of uranium were still incomplete, and that the potential hazards of working with ether were still present.

It was possible to get higher recoveries with a newer solvent discussed in Section II, Part 4. (Based on a uranium value of $10.00 per pound, an additional 0.1 per cent of recovery is equivalent to one cent per pound in processing cost.)

Production And Costs

Production and cost details are contained in Section IV.

Related Documents

The following numbers refer to bibliography entries which are the principal sources for more detailed information on the development of the continuous ether-extraction process described in this subsection: 5, 8, 10, 19, 41, 67, 87, 93, 104, 122, 155, 176, 186, 190, 199, 201, 211, 222, 223, 224, 225.

Additional references may be found in some of the sources indicated above.

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Section II
Part 4

DEVELOPMENT OF THE TRIBUTYL PHOSPHATE - HEXANE PROCESS FOR URANIUM PURIFICATION

The first plant-scale uranium purification system to use tributyl phosphate (TBP) in hexane as the extractant was started at Weldon Spring, Mo., in June, 1957. The start-up culminated seven years of research and development by Mallinckrodt Chemical Works and marked a major advance in uranium processing.

Background

As early as 1950, seeking an improvement over the use of ether as an extractant, Mallinckrodt began to study other potential solvent extraction systems. Earlier work at Iowa State University and at Oak Ridge National Laboratory had indicated the possibility that TBP might be used. In October, 1951, Mallinckrodt's researchers completed their initial investigations and concluded that diluted TBP would be a considerably better extractant than ether.

Development of the TBP-kerosene system, which was used at Fernald, Ohio, beginning in January, 1954, was based on this original work at Mallinckrodt.

Reasons For Selection Of Diluted TBP As The Extraction Solvent

The two chief reasons for the selection of diluted TBP to replace ether as the extractant in uranium purification were:

(1) Safety. A system using TBP, which is stable to high concentrations of nitric acid at temperatures normally encountered in the purification process, is relatively free of the potential hazards present in ether systems.

(2) Economies. Lower capital and operating costs are possible in a purification system using diluted TBP. TBP, with its high selectivity for uranium, gives considerably better recoveries from ores than ether. Furthermore, adequate purity can be achieved through the proper use of a single extraction cycle.

Need For Dilution

If TBP were used as a solvent in the pure state, certain of its physical properties would create problems during extraction because:

- The density of TBP is so nearly identical to that of water that good phase separation in liquid-liquid extraction is difficult. In addition, this property of TBP would interfere with the operation of a continuous system.

- The high viscosity of TBP reduces the efficiency of extraction and promotes the formation of stable emulsions when agitated with aqueous solutions.

- The high degree of stability of the undiluted uranyl nitrate-TBP complex is such that re-extraction of the uranium portion becomes difficult.

Dilution with an inert organic liquid minimizes these disadvantages of TBP by lowering density and viscosity and improving phase separation and mass transfer. Mallinckrodt's early investigations revealed that the lower paraffin hydrocarbons would serve as suitable diluents. This was substantiated by the favorable results achieved with the use of kerosene as the diluent for the Fernald plant, and by the improved operation using hexane as the diluent for the Weldon Spring plant.

The Basic TBP Purification Process

The basic purification process at Weldon Spring involves four major steps.

First, nitric acid is used to digest the ore concentrate. Then, the resulting slurry is extracted with the diluted TBP. In the third step, small amounts of water are used to wash impurities from the uranium-bearing extract. Finally, the clean uranium is re-extracted into water, and the solvent is recycled.

Figure II-4.1 presents a generalized flow sheet illustrating the process.

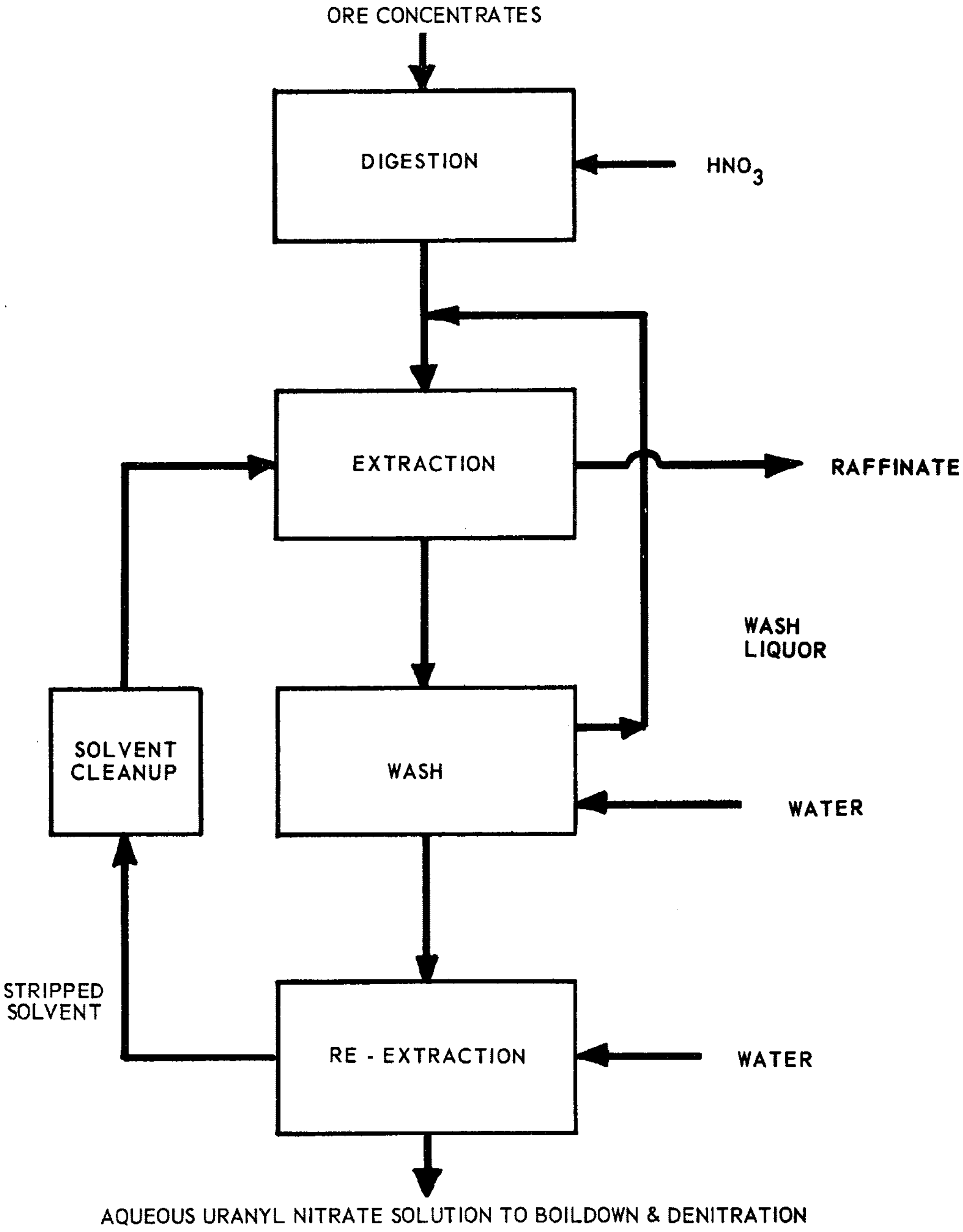

Figure II-4.1 THE BASIC TBP PURIFICATION PROCESS involved four major steps: digestion, extraction, washing and re-extraction.

Pilot-Plant Development

In 1954, when design work for the Weldon Spring plant was started, the experience with the TBP-kerosene process at Fernald indicated that some improvements might be possible.

Subsequently, Mallinckrodt carried out an extensive pilot-plant development program which resulted in four major advancements over the TBP-kerosene process. They were:

(1) Selection of hexane as the diluent instead of kerosene.

(2) Design and development of pumper-decanters as the primary extraction contactors instead of pulse columns.

(3) Design and development of "high-capacity" pulse columns with advanced plate configuration and changed plate spacing for more efficient re-extraction.

(4) Use of a slurry feed with higher uranium and lower nitric-acid concentrations.

The Use Of Hexane As A Diluent

Although kerosene had been used with favorable results at the Fernald plant, Mallinckrodt -- in its laboratory and pilot-plant research -- sought an improved diluent for use in the Weldon Spring facility.

Hexane was selected because it offered the following advantages over kerosene.

- The lower density and viscosity of hexane resulted in improved phase separation and consequently permitted greater capacity in extraction contacting equipment.
- The lower viscosity of TBP in hexane improved mass transfer.
- Hexane offers greater stability than kerosene in the presence of nitric acid.

In addition, because the lower viscosity of hexane results in less entrainment of impurities during primary extraction, it produces a purer product. Furthermore, because hexane is more volatile than kerosene, it can be completely removed by distillation from aqueous streams.

Development Of Pumper-Decanters

In early experiments, spray columns, jet-mixer columns, pulse columns, and various types of mixer-settlers were tested for use in the primary extraction step. The unit that proved most satisfactory was the pumper-decanter.

The pumpter-decanter is a type of mixer-settler in which the organic and aqueous phases are first mixed externally in a centrifugal pump and then allowed to separate in a tank or decanter.

The chief difference between the pumper-decanter and conventional mixer-settlers is that the pumper-decanter has provisions for internal solvent recycle. This permits a high ratio of organic to aqueous in the mix pump, greatly increasing the solids-handling capability of the unit.

Evolution of the pumper-decanter design was based on the following experimental observations:

- To obtain the good phase contact required for high extraction efficiency, vigorous agitation is necessary; however, when agitated vigorously, the TBP-hexane and the aqueous slurry have a tendency to emulsify.
- By maintaining a high solvent to aqueous ratio (at least 10/1), the emulsification problem is reduced substantially.
- When the system is operated with the less viscous fluid (the solvent) as the continuous phase, considerably less emulsification results.
- After mixing, separation of the two phases is promoted when the solvent (continuous) phase is in rotary motion.

Operation of the pumper-decanter system designed by Mallinckrodt is illustrated schematically in the diagram shown in Figure II-4.2.

A general view of the pumper-decanter equipment as it appeared in the plant is shown in the photograph in Figure II-4.3.

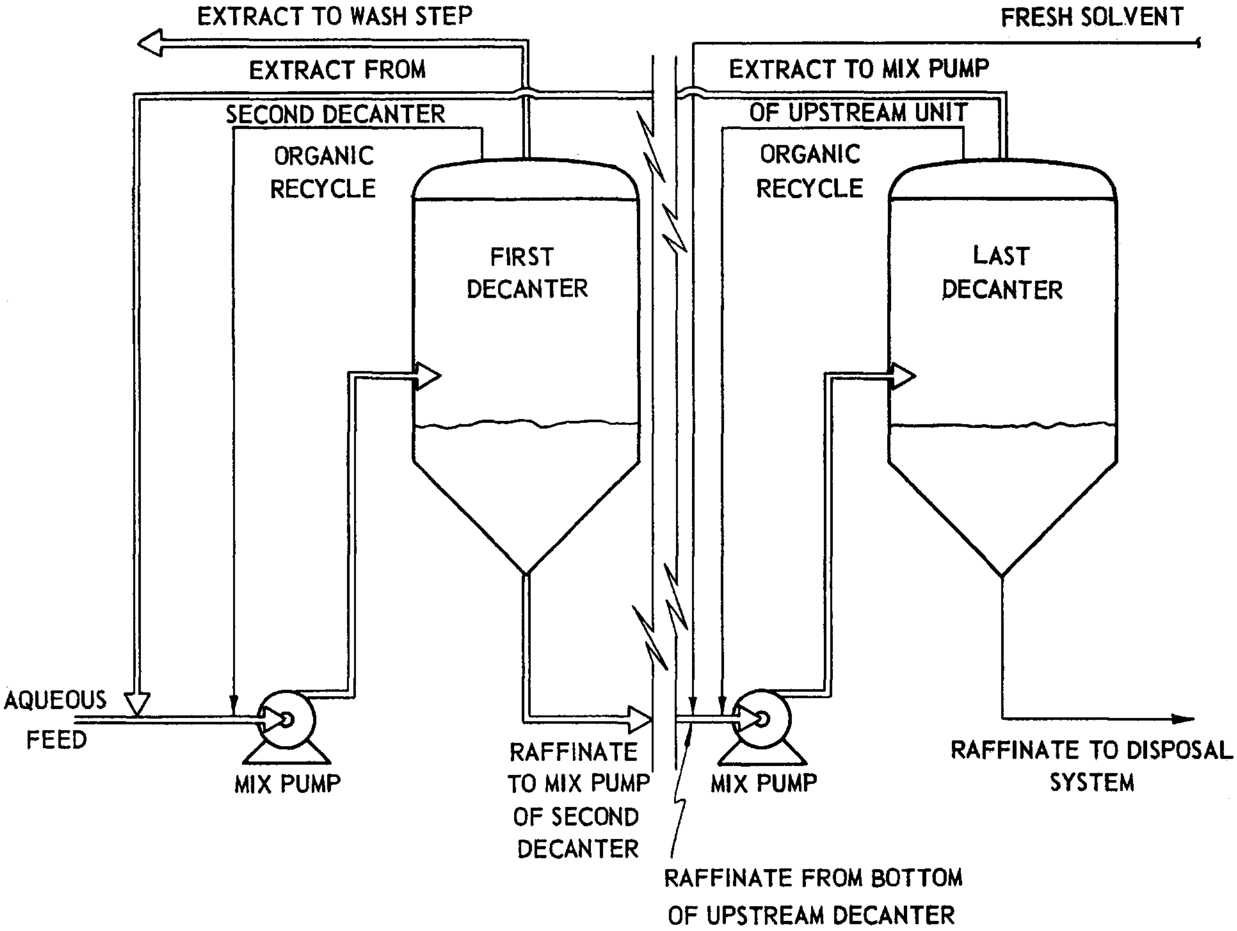

Figure II-4.2 SCHEMATIC FLOW DIAGRAM OF PUMPER-DECANTERS -- end stages.

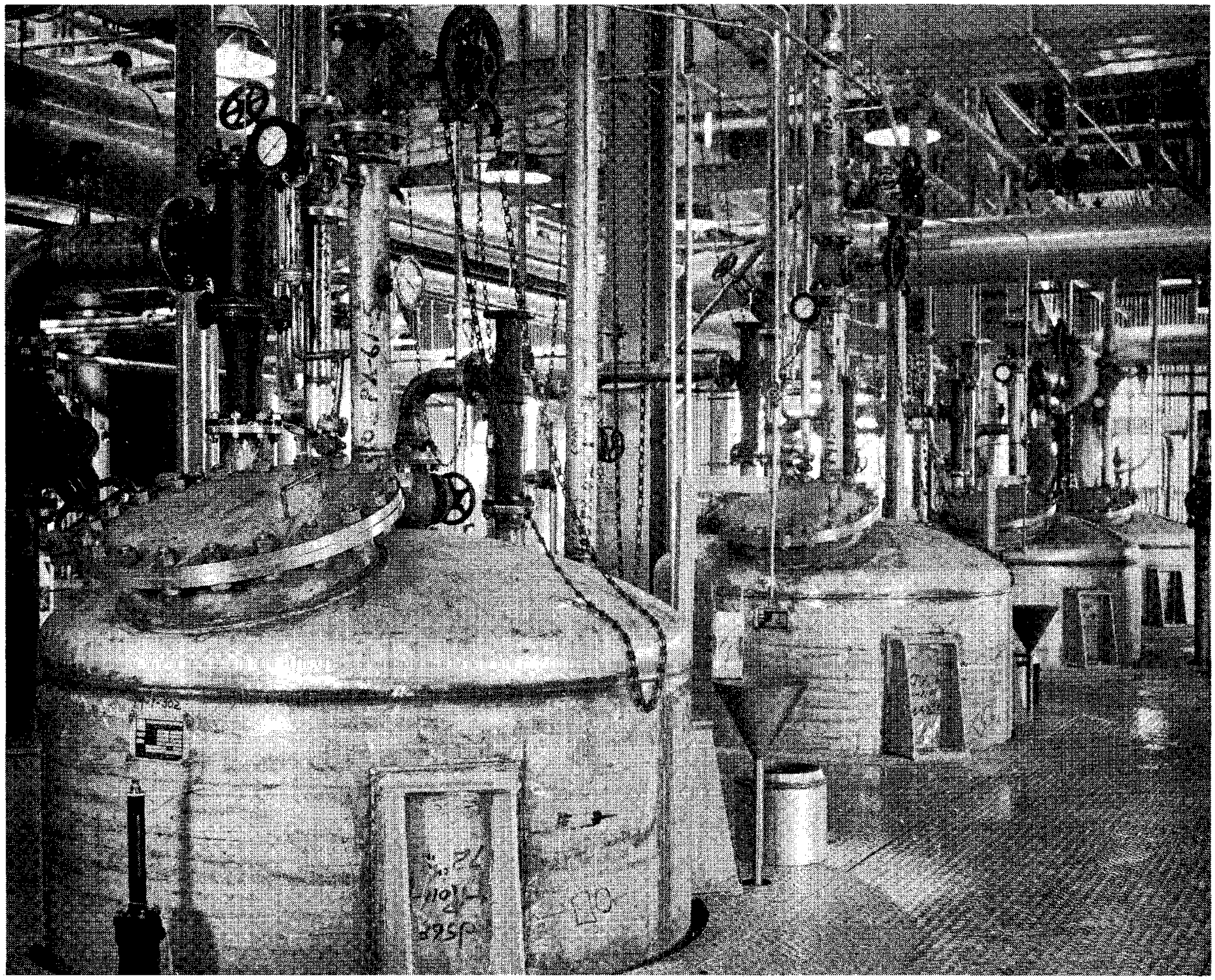

Figure II-4.3 PUMPER-DECANTER EQUIPMENT used in TBP-Hexane extraction is shown here as it appeared in the plant.

Development of High Capacity Pulse Columns

Pumper-decanters and pulse columns were compared in the pilot plant for possible use in scrubbing and re-extraction. Although the pumper-decanters performed satisfactorily, scrubbing and re-extraction operations required a large number of stages. For this reason, pulse columns were selected as the more economical equipment for installation in the plant.

In all of the intitial experiments using pulse columns, instability of operation occurred when the solvent was the continuous phase. Therefore, the conditions were reversed. The columns were operated with the aqueous phase continuous, and more stable operation was achieved.

For the plant installation, a single column was chosen for the washing operation. Two columns operating in parallel were installed for the re-extraction operation.

The use of hexane as the diluent, and the increase in the capacity of the pumper-decanters made it desirable to increase the extraction capacity of the pulse columns to achieve a balance between extraction and re-extraction. Since the two re-extraction columns could accommodate more transfer stages than were actually required, an attempt was made to modify the internal design of the column to permit higher extraction capacity at the expense of a small decrease in extraction efficiency.

Extraction capacity of a pulse column is ordinarily limited by the degree of emulsification. Since emulsification is the result of excessively fine dispersion of one phase within the other, any increase in extraction capacity would require decreasing the degree of dispersion of the organic within the continuous aqueous phase. This was ultimately achieved by developing an internal column configuration involving plates with: large (3/8 inch) hole diameters; relatively large (33%) free area; Teflon coating, and wide (4-inch) plate spacing.

These developments resulted in high-capacity re-extraction pulse columns. The columns have nearly three times more extraction capacity, using TBP-hexane, than other pulse columns used in similar operations with TBP-kerosene. In addition, the capacity of the columns also was increased by 400 to 500 per cent as a result of the developments.

Development Of The High-Uranium, Low-Acid Flowsheet

Until the time of the TBP-hexane pilot-plant investigations, the feed concentration ordinarily used in uranium-extraction operations was 200 grams of uranium per liter in 3<u>N</u> nitric acid. When the pumper-decanters -- with their ability to handle feed slurries high in solids content -- were developed, it became possible to use a feed slurry with an increase in the uranium and a decrease in the acid concentrations. Therefore, a high-uranium, low-acid flowsheet was developed.

The high-uranium, low-acid flowsheet offered the following advantages over previous systems:

- It substantially increased production capacity. The chief limit to the capacity of the pumper-decanters is the volume of the aqueous stream that they can handle. Consequently, an increase in concentration of uranium in the feed results in an almost proportional increase in uranium-extraction capacity.

- The low-acid feed eliminated the need, and consequent expense, for nitric-acid recovery from the raffinate (discard) stream. With low-acid concentration, it is more economical to neutralize and discard the nitrates than to recover them by boildown and concentration.

- Savings were achieved as a result of the smaller quantities of nitric acid required.

- The low-acid stream decreased corrosion problems throughout the system.

- Better control of impurities was provided. The high concentration of uranium in the feed permitted a higher degree of uranium saturation of the solvent and a consequent reduction in the amount of impurities carried forward in the primary extract.

Scale-Up To A Plant System

The plant design was based on operating conditions derived from the laboratory and pilot-plant work described above.

In June, 1957, the full-scale refinery started operation, and all elements performed satisfactorily. The plant operated without major difficulties from initial start-up until the Weldon Spring facility was closed in 1966.

Conclusions

Development of the tributyl-hexane process -- including Mallinckrodt's laboratory research, pilot-plant work, plant scale-up, and ongoing improvements -- made Weldon Spring the most advanced uranium-extraction plant in existence.

Major economic benefits were derived from the technical and operational development and the advancements that Mallinckrodt achieved.

Because of the comprehensive and intensive nature of the pilot-plant work, the plant that Mallinckrodt designed was a flexible one. Thus, even though basic designs and sizes of plant equipment were frozen at an early stage, it was possible, as a result of continuing developments and systematic studies of operating conditions, to approximately triple the capacity of the plant without additional major capital expense.

The resulting economies in plant operation contributed substantially to allowing the AEC to close down the older and by-then obsolete ether process refinery. Savings achieved in direct operating costs alone totaled approximately $1 million annually.

Production And Costs

Production and cost details are contained in Section IV.

Related Documents

The following numbers refer to bibliography entries which are the principal sources for more detailed information on the development of the TBP - hexane uranium purification process described in this subsection: 10, 13, 16, 41, 57, 67, 81, 83, 89, 94, 118, 128, 146, 151, 157, 158, 173, 186, 187, 199, 201, 211, 222, 223, 224, 227.

Additional references may be found in some of the sources indicated above.

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Section II
Part 5

DEVELOPMENT OF THE POT PROCESS
FOR
CONVERTING URANYL NITRATE TO ORANGE OXIDE

In the spring of 1942, as a major part of its initial work in connection with the Manhattan (atomic bomb) Project, Mallinckrodt Chemical Works developed the pot process for denitration of uranyl nitrate to produce uranium trioxide (UO_3, or orange oxide). With continuing refinements, the process remained the standard denitration method at Mallinckrodt's St. Louis-area operations until the Weldon Spring activities were terminated.

Background

In April, 1942, Mallinckrodt assigned research and engineering personnel to seek solutions to two major technical problems in efforts to produce the highly purified uranium compounds needed by Dr. Arthur Holly Compton and his colleagues for use in the experimental nuclear reactor at the University of Chicago.

One of the problems was the development of a safe, commercial ether-extraction process. (For details, see Section II, Part 2.) The other problem was to develop a practical process for converting the uranyl nitrate product into a satisfactory uranium-trioxide product.

The effort to develop a uranium-trioxide production process was started at Mallinckrodt's Jersey City, N.J., facility. The Company already was producing red mercuric oxide there by a process similar to the one being considered for use in converting uranyl nitrate to UO_3.

In the mercuric-oxide production at Jersey City, an aqueous, acid solution of mercuric nitrate was heated and stirred simultaneously in small, stainless-steel kettles. The liquid gradually evaporated, and, as the temperature rose, the mercuric nitrate decomposed. Nitric-acid fumes were driven off by the heat, and the mercuric oxide was formed as the remaining product.

The Basic Batch Denitration Process

The method that Mallinckrodt decided to investigate for producing orange oxide was similar to the mercuric-oxide process at Jersey City.

The process involved thermal decomposition of uranyl nitrate in pots. A highly purified uranyl nitrate solution is heated and agitated in specially fabricated stainless steel kettles. The heat drives off the oxides of nitrogen and water, and, in the process, the solution is ultimately transformed to uranium-trioxide powder.

A sketch of the type of denitration pots used is presented in Figure II-5.1.

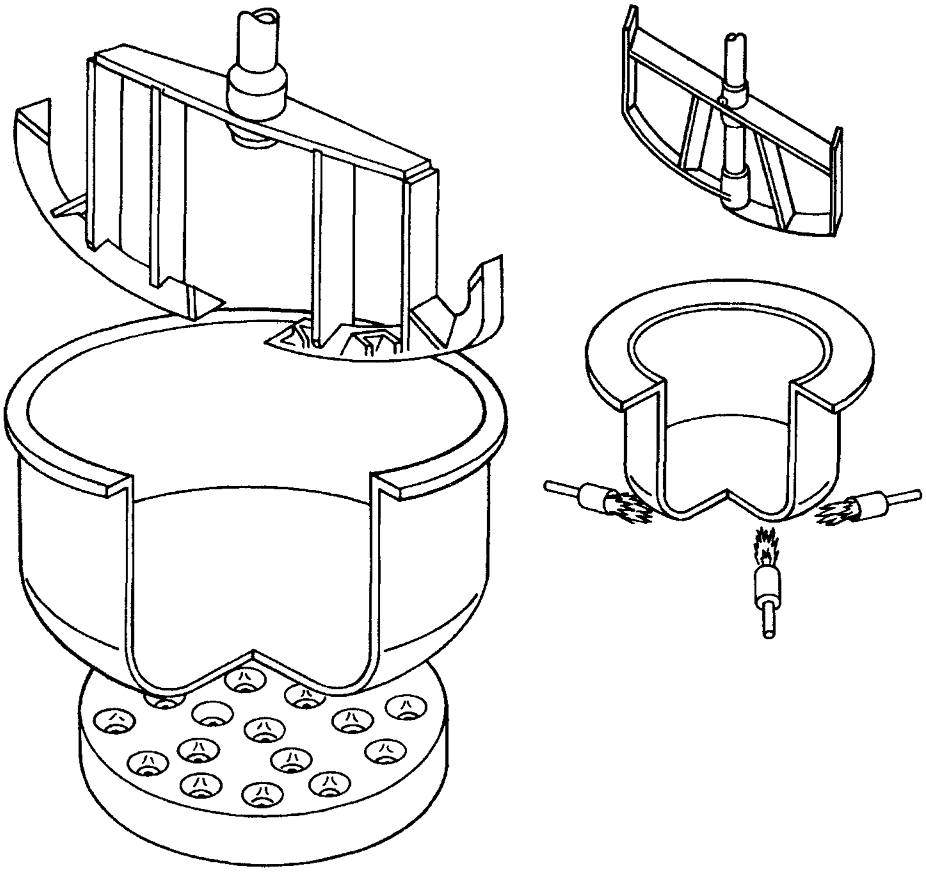

Figure II-5.1 SPECIALLY FABRICATED STAINLESS STEEL POTS were used in the batch denitration process.

Initial Experimental Work In Jersey City

Mallinckrodt research personnel initially tried thermal decomposition of uranyl nitrate in the laboratory, and the experiment proved successful. The next step was to try the process in the plant.

The University of Chicago group arranged for several thousand pounds of black uranium oxide to be delivered to the Mallinckrodt plant in Jersey City. The first step in the plant-scale experiment was to dissolve the black uranium oxide in nitric acid. The resulting solution was then filtered into the stainless-steel kettles. Next, the coal fire which heated the kettles was started, and the stirrer was turned on.

As the heating and stirring proceeded, the water evaporation, the thickening of the mixture in the pots, and the brown, nitrogen-oxide fumes coming off indicated that the anticipated decomposition of uranyl nitrate was occurring. As the reaction continued, the material in the kettles became doughy and ultimately turned into a bright orange powder -- the sought-after uranium-trioxide product.

After the experiment was repeated twice, each time with similar results, the Mallinckrodt researchers concluded that the process could be used for plant-scale production of the necessary uranium trioxide.

Problems In Acquiring And Installing Equipment For The St. Louis Plant

After the basic process had been developed and proved in Jersey City, the next problems that Mallinckrodt faced concerned the acquisition and installation of the equipment needed to establish the uranium-trioxide production plant in St. Louis.

To avoid using coal fire, which was both cumbersome and hard to control, to heat the reaction pots in St. Louis, Mallinckrodt built gas-fired Dutch ovens. The design was based on that of the oil-fired Dutch ovens used to heat the mercuric-oxide reaction pots at the Company's plant in Montreal, Canada.

Mallinckrodt's personnel in Jersey City tried to purchase stainless-steel kettles for installation in St. Louis; however, stainless-steel was practically unavailable at the time. Even for high-priority orders, a minimum of three to six months was required for delivery. To get the job done in time to meet the needs of the Chicago group, Mallinckrodt decided to dismantle the red-oxide plant in Jersey City and ship the equipment on flatcars to St. Louis.

Another problem became evident when the equipment arrived in St. Louis. The shipment included 220-volt, two-phase, gearhead motors for which there was no appropriate electrical power at the St. Louis plant. This obstacle was overcome when, through the resourcefulness of Mallinckrodt's personnel, special transformers were located which converted the three-phase power of the St. Louis plant to two-phase power.

Activities During Plant Start-Up

After the installation of the necessary equipment was completed, the St. Louis plant was ready for start up.

At first, the plant operated without incident. The equipment performed satisfactorily and the denitration process proceeded as anticipated. However, as the reaction mixtures in the pots began to thicken, a mechanical problem developed: the agitators began to bind. As the binding continued, the load on the motors that drove the agitators increased until the motors finally tore loose carrying with them large chunks of the concrete floor to which they had been bolted.

The damage was repaired, and to preclude further occurrences, Mallinckrodt installed stronger braces for the motors and agitators. In addition, the blades of the stirrers were changed to prevent binding. After these changes were incorporated, the plant operated without further significant technical difficulties.

One of the early denitration kettles is shown in Figure II-5.2.

Ongoing Improvements

The process initially developed by Mallinckrodt in 1942 continued to be the basic denitration technology used for production of UO_3 throughout the entire history of feed-material processing in the St. Louis/Weldon Spring area.

A number of improvements and refinements were made, however, over the years. When the uranium processing operation moved to Mallinckrodt's Destrehan Street plant, three major changes were incorporated. They were:

(1) Installation of burners directly under the pots to replace the Dutch ovens which had been used at the main St. Louis plant. The result was more efficient heating.

(2) Installation of a more effective and efficient nitric-acid recovery system.

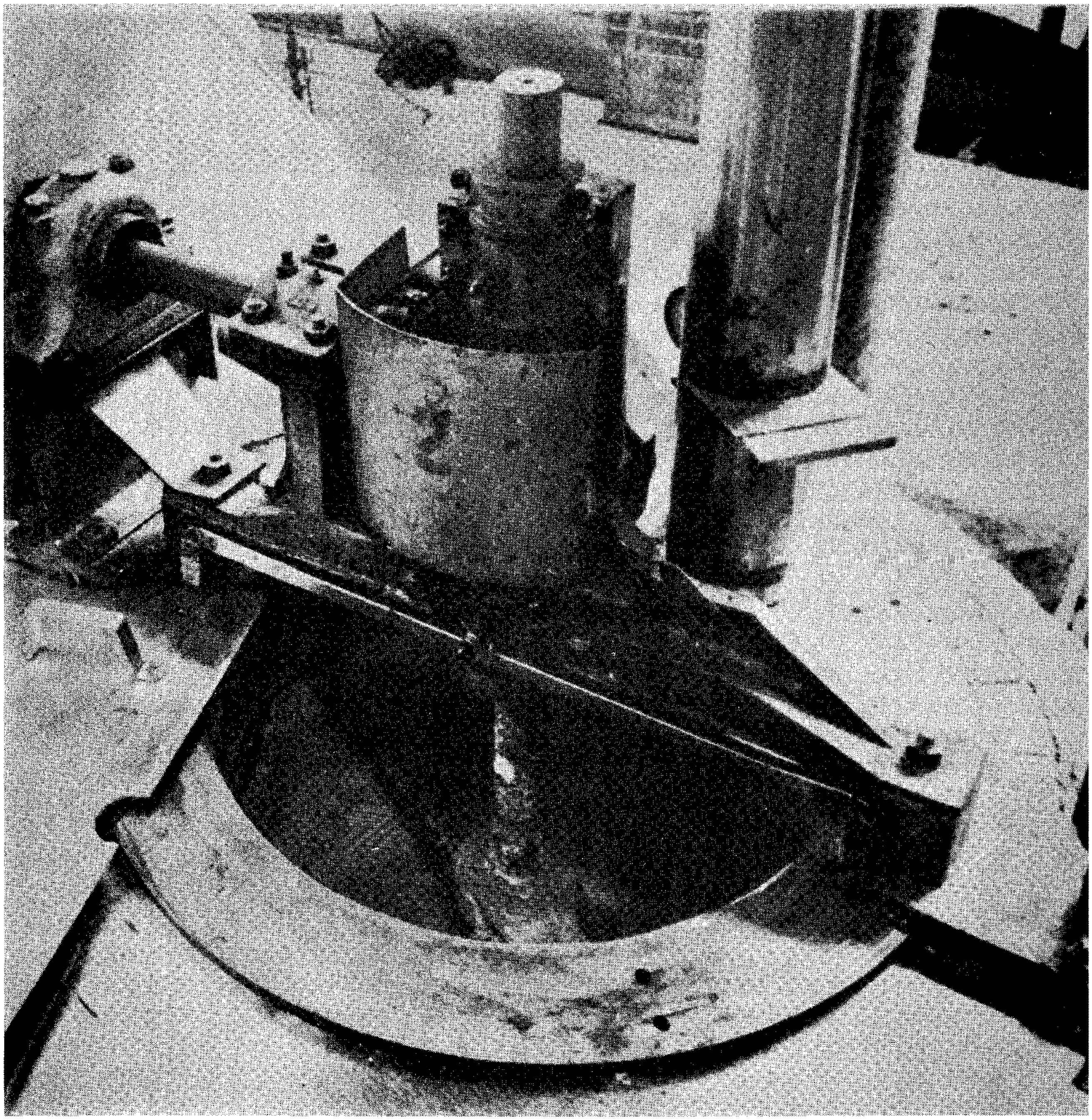

Figure II-5.2 DENITRATION KETTLES of this type were used in early production of orange oxide.

(3) Replacement of the manual unloading technique, which had been used at the main plant, with a pneumatic unloading system. The pneumatic unloading operation is shown in Figure II-5.3.

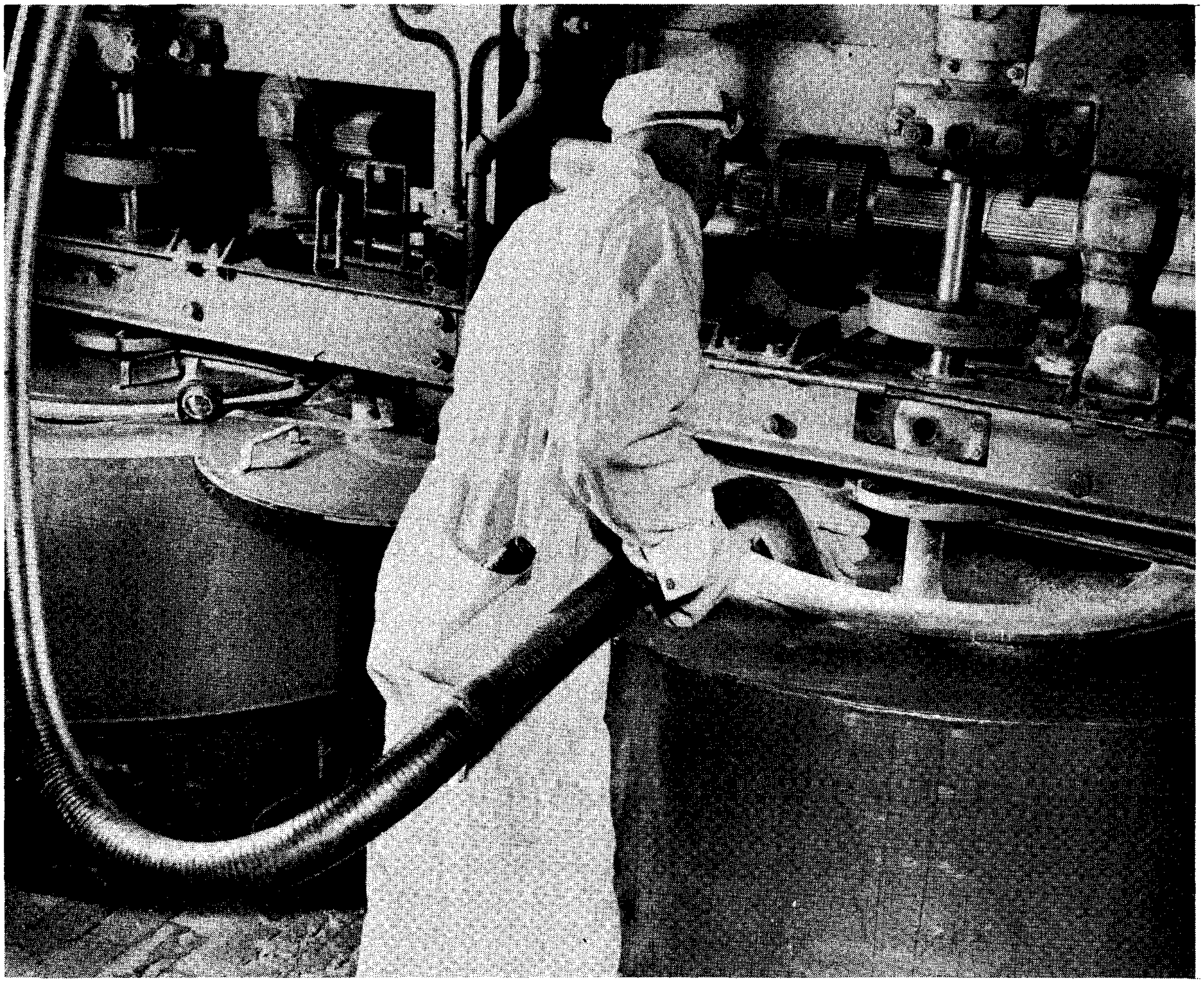

Figure II-5.3 PNEUMATIC UNLOADING, which replaced manual unloading of the denitration pots, was one of the improvements Mallinckrodt made.

Five other major changes were made to the denitration system at the Weldon Spring plant. They were:

(1) Installation of wider and deeper pots to increase production capacity without increasing the number of operators.

(2) Incorporation of a more sophisticated flame-failure safety system.

(3) Installation of a remote-controlled ignition system which was safer and more convenient than the manual ignition technique which had been employed at Destrehan Street.

(4) Incorporation of a programmed temperature-control system which resulted in improved product of a more uniform quality.

(5) Installation of an improved fume-control system and a better dust-collection system.

Conclusions

As it did in developing a safe, commercial ether extraction process, Mallinckrodt -- operating under the handicaps of time pressures and war-time shortages of equipment and materials -- developed a successful, tonnage-scale process for converting uranyl nitrate into satisfactory-grade uranium trioxide.

The process's chief advantages were that it worked well, and it was simple. That the basic process, with improvements, was used for nearly a quarter of a century is evidence of its practicality and reliability.

The process's economical disadvantages and the dusty operations, both inherent in its being a batch process, and the possibility of improving the quality of the product led to Mallinckrodt's development of the fluid-bed (continuous-process) denitration system which is discussed in detail in Section II, Part 6.

Production And Costs

Production and cost details are contained in Section IV.

Related Documents

The following numbers refer to bibliography entires which are the principal sources for more detailed information on the history of the pot denitration process described in this subsection: 8, 10, 11, 13, 16, 19, 41, 57, 67, 70, 83, 172, 173, 177, 178, 186, 199, 201, 211, 212, 222, 223, 224, 225.

Additional references may be found in some of the sources indicated above.

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Section II
Part 6

DEVELOPMENT OF THE FLUID-BED DENITRATION SYSTEM

Mallinckrodt Chemical Works' activities in developing a fluid-bed process for denitration of uranyl nitrate to produce uranium trioxide (UO_3, orange oxide) were started at Weldon Spring in the late 1950's.

Background

The fluid-bed project was one of a number of attempts throughout the country to develop a continuous process that would be a more economical, more efficient, and more effective method of denitration than the established pot process.

The pot process, which is discussed in detail in Section II, Part 5, is a batch operation. It involves thermal decomposition of a highly purified, uranyl nitrate solution in specially fabricated, stainless steel pots which are heated by gas burners. The uranyl nitrate solution is heated and agitated in the pots. Water is driven off and nitrates decompose gradually. The solution is transformed first to a dough, and finally to uranium-trioxide powder. A pneumatic conveying device is employed to remove the powder from the pots for subsequent use in green-salt production.

This process works well. It is a simple operation with the added advantage of simplicity in equipment design. However, the process also has several disadvantages, including:

(1) Direct operating costs are high. The process requires many operators to run a large number of pots.

(2) Costs are high for equipment maintenance. The stainless steel pots have a high failure rate and require frequent replacement and/or repair.

(3) Control of temperature and other technical factors influencing product quality in the production process is limited.

(4) The system, an open one, presents some health hazards associated with orange-oxide dust and fumes.

Potential For Economies And Improved Quality

Early work at Argonne National Laboratory indicated that denitration in a fluid-bed reactor might be a feasible alternative to the pot process.

With fluid-bed denitration, it seemed likely that significant savings could be realized in the cost of equipment, operations, and maintenance. Furthermore, the fluid-bed reactor would afford better control over technical factors, such as temperature, which influence the quality of material produced during denitration.

The Fluid-Bed Reactor

A fluidized bed is a bed of solid particles supported by a fluid flowing through it at a specific velocity in an upward direction. In uranium production, gas is used as the fluid. The bed is contained in a shell, and to allow for expansion, the bed is not restricted on top.

When the gas passes through at the proper velocity, it causes the bed of solid particles to expand. The particles become suspended in the gas, and instead of resting upon one another, they separate and move about freely, assuming the characteristics of a fluid.

Development Of The Fluid-Bed Processing Technique

Initial development of material processing using fluid-bed reactors occurred in the United States in the petroleum industry during the early 1940's in connection with efforts to establish a better process for producing gasoline and oil from crude petroleum.

The petroleum industry's work made it apparent to observers in other process industries that fluid-bed reactors offered a number of attractive features.

Their principal advantages included: (1) excellent temperature control, (2) continuity of operation, (3) high heat transfer rates, and (4) excellent fluid-solid contact. All of these features have significance in uranium processing, and the first three are especially important in thermal denitration of uranyl nitrate.

Initial Development Of Fluid-Bed Denitration

In 1954, Argonne National Laboratory, seeking a continuous process as an alternate to the established batch method of denitration, initiated development work on fluid-bed denitration. As part of the project, Argonne built and operated a small, laboratory-scale fluidized-bed denitration reactor.

The Process

The fundamental process involved in fluid-bed denitration is illustrated in Figure II-6.1.

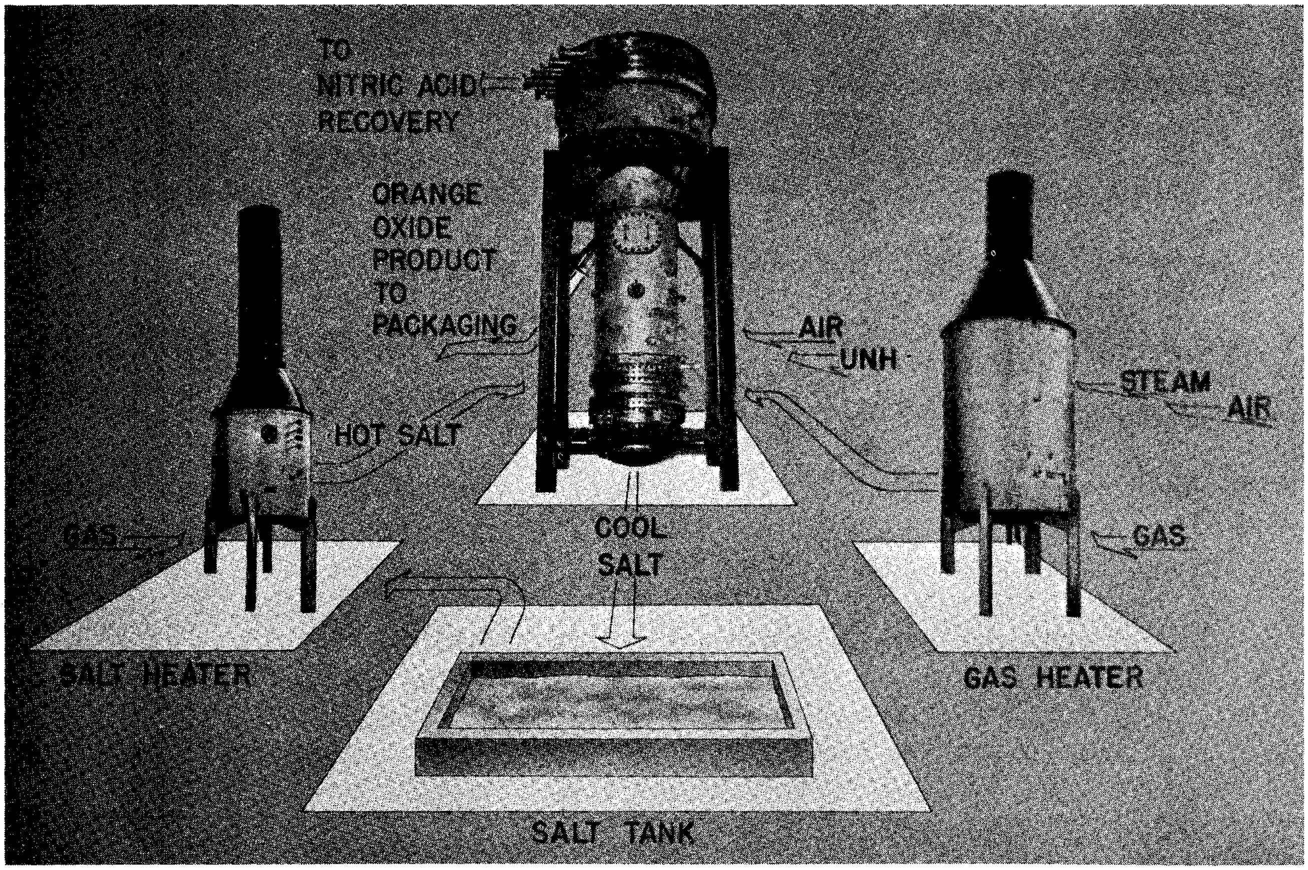

Figure II-6.1 BASIC FLUID-BED DENITRATION PROCESS.

In the first step in fluid-bed denitration, a bed of uranium trioxide particles is fluidized by a gas (usually air and/or steam) and heated to 500°F-700°F. At that temperature range, evaporation and thermal decomposition of uranyl nitrate occurs. A uranyl nitrate solution (UNH, uranyl nitrate hexahydrate) is then sprayed into the fluid-bed reactor.

When individual droplets of the spray enter the fluid-bed reactor, they are heated by contact with individual uranium-trioxide particles moving about freely in the denitrator. The heat causes rapid evaporation and thermal decomposition of the droplets and transforms them into uranium-trioxide solids and nitric acid fumes.

The chemical reaction that occurs is identical to the reaction in the pot process. Heat applied to the uranyl nitrate hexahydrate removes water and nitrates as gases, and produces uranium trioxide as the remaining solid.

Unlike the pot method, fluid-bed denitration has the advantage of being a continuous process. As denitration of the uranyl-nitrate liquor fed into the reactor occurs, the volume of orange oxide in the bed increases. The excess over the original volume of UO_3 in the bed represents the product. It is continuously removed from the reactor at a regular rate, being replaced by newly produced orange-oxide product.

Initial Pilot-Plant Development In St. Louis

The encouraging results of Argonne's laboratory scoping efforts prompted an extensive, full-scale research and development program at the Mallinckrodt facilities in St. Louis. The ultimate, long-range objective of the program was to develop a practical fluid-bed denitration system that would be successful on a plant scale.

In 1957, a pilot-scale fluid-bed reactor, somewhat larger than the first small-scale reactor at Argonne, was installed at Mallinckrodt's Destrehan Street facility in St. Louis. A sketch of the reactor is shown in Figure II-6.2.

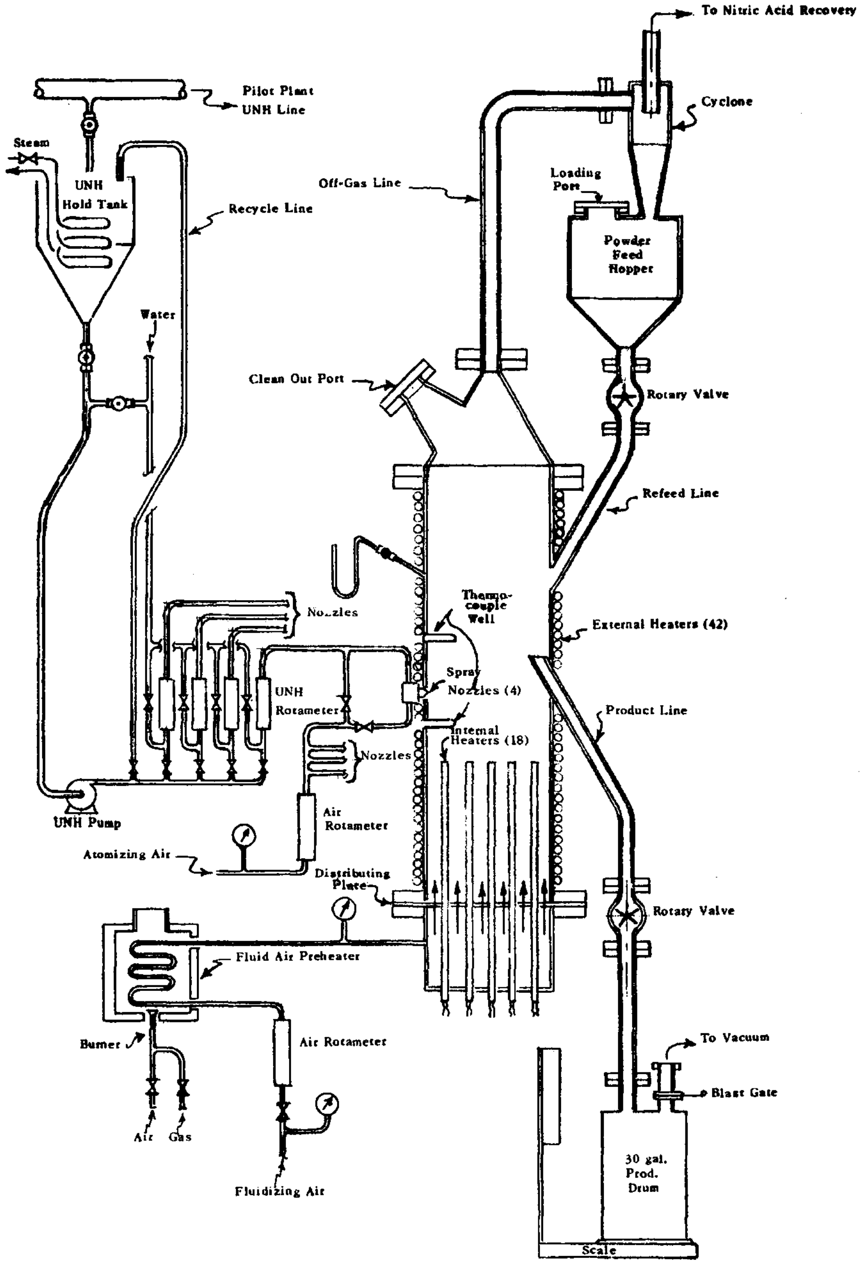

Figure II-6.2 DESTREHAN STREET PILOT-PLANT FLUID-BED DENITRATOR.

The Destrehan Street pilot-plant fluid-bed denitrator advanced the design concept of the Argonne denitrator by incorporating several improvements. The major ones are listed below:

(1) The Argonne reactor had been heated with external heaters only. The Destrehan Street unit employed 18 internal, electrical bayonet heaters in addition to 42 external heaters.

(2) At Destrehan Street, the off-gas system used a cyclone instead of sintered metal filters, which had been used at Argonne.

(3) At Argonne, the product had been withdrawn from the bottom of the bed. With the Destrehan Street reactor, the product discharged through an overflow opening at the top of the bed.

Pilot-Plant Work At Weldon Spring

In 1958, a new pilot-plant fluid-bed denitrator was put into operation at Weldon Spring. Figure II-6.3 illustrates the main features of the equipment as originally built.

The Weldon Spring unit was considerably more versatile than the one at Destrehan Street and permitted greater flexibility of operation in terms both of operating conditions and production rate.

One of the major differences between the new Weldon Spring denitrator and the one at Destrehan Street was the method of heating. In the Weldon Spring reactor, heat was supplied by molten salt circulating through 30, one-inch, stainless-steel bayonet tubes located inside the reactor. The Destrehan Street design had used external and internal electrical heaters. To provide better temperature control, the electrical heaters were replaced in the new design with the molten-salt system.

The original Weldon Spring pilot-plant fluid-bed denitrator was later modified as shown in Figure II-6.4.

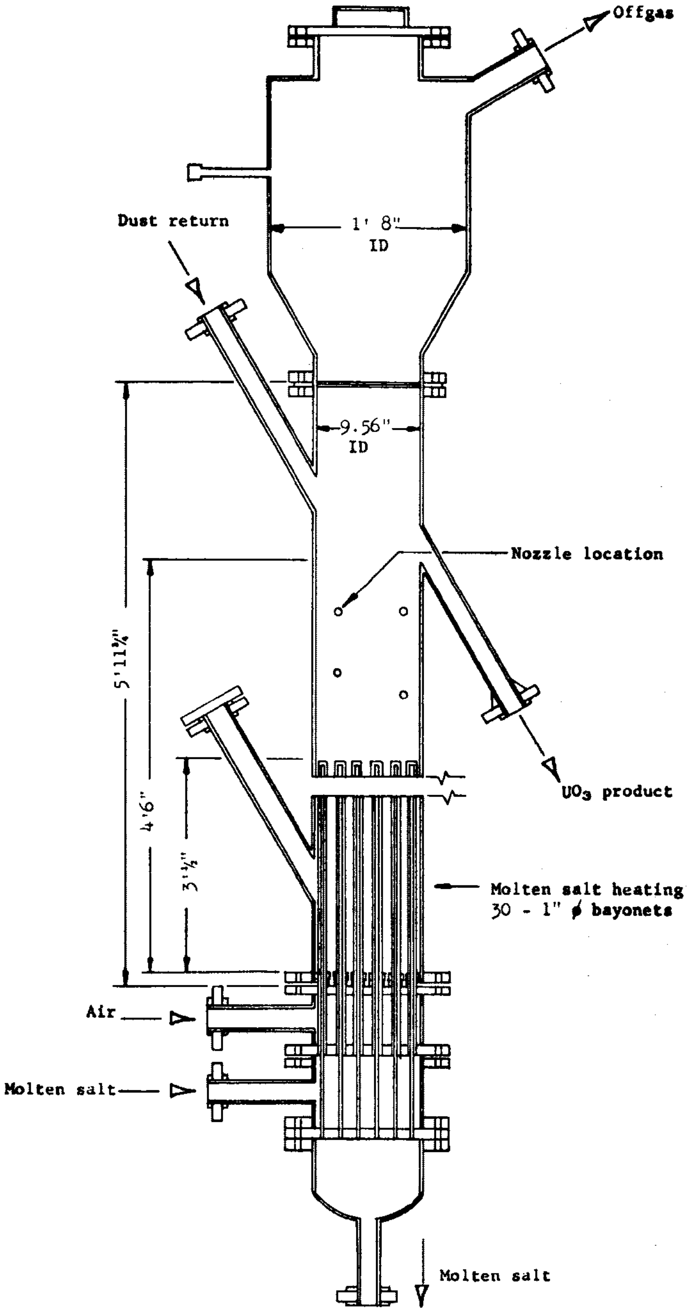

Figure II-6.3 ORIGINAL WELDON SPRING PILOT-PLANT FLUID-BED DENITRATOR.

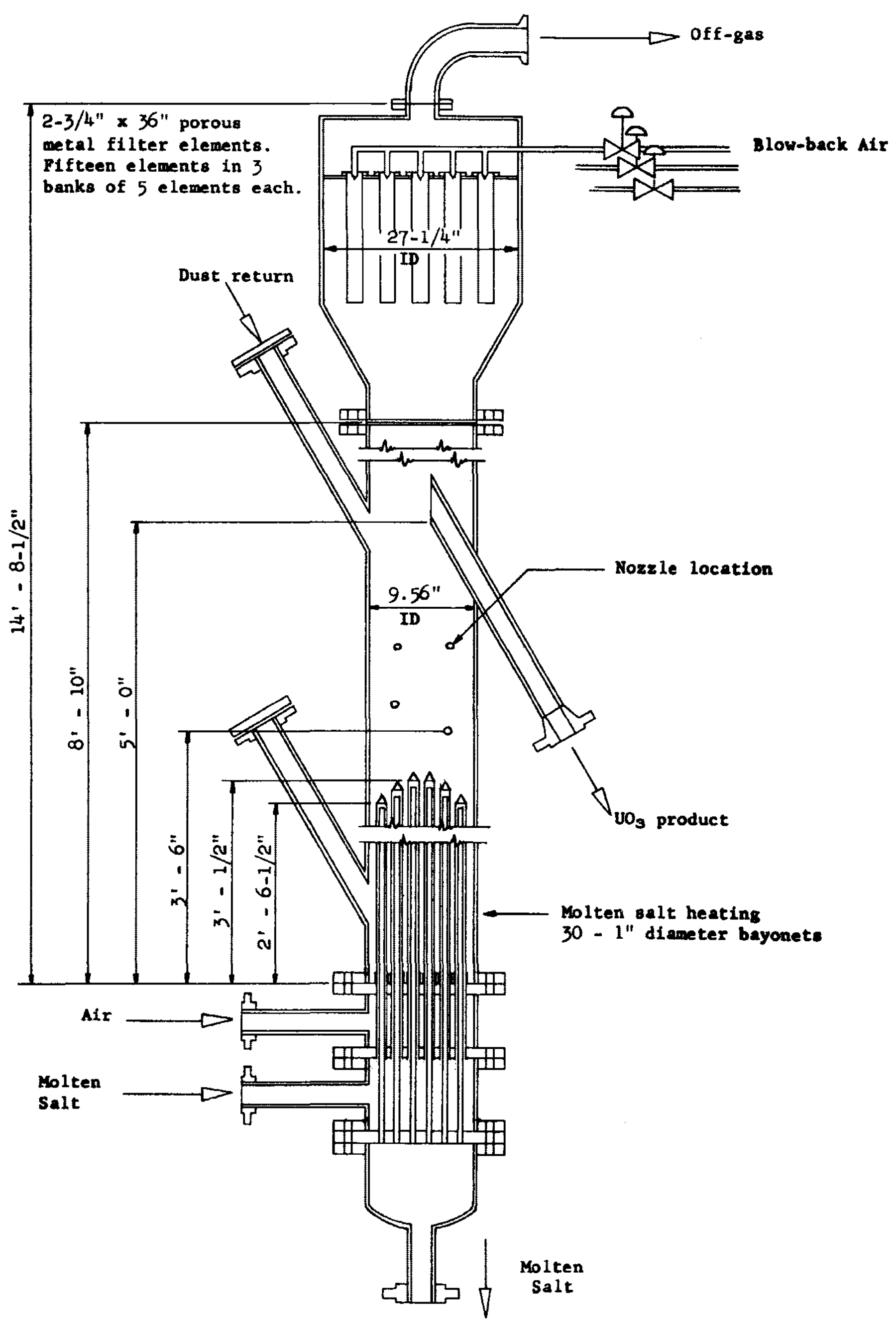

Figure II-6.4 MODIFIED WELDON SPRING PILOT-PLANT FLUID-BED DENITRATOR.

The chief differences between the modified equipment and the original equipment were:

(1) A stainless steel, steam-jacketed filter and a blow-back device were installed in the modified denitrator in place of the cyclone separator and refeed system used originally. The new filtering system consisted of three filter banks, each of which had five porous-metal filter elements. The filter banks were automatically back-blown at predetermined intervals with pre-heated air.

(2) A new molten-salt heating system with bayonet tubes of varying lengths was installed in place of the original molten-salt system which used tubes of equal length.

Development Of A Plant-Scale Fluid-Bed System At Weldon Spring

In May, 1964, a plant-scale fluid-bed denitration system was installed at Weldon Spring. Its design was based on the experience and knowledge gained from the pilot-plant work at Destrehan Street and at Weldon Spring.

Start-up activities, which began in June, 1964, were interrupted by special work elsewhere in the plant. They were resumed in December, 1964. From then until April, 1966, the start-up work involved additional development.

At first, it was found that the uranium-trioxide product of the plant-scale system was not reactive enough for subsequent hydrofluorination to metal-grade green salt. The quality of the orange oxide produced was, however, ideal for use in gaseous diffusion plants.

To solve the reactivity problem, further analytical work was conducted, and a planned experimental program was executed. This additional research and development work resulted in a practical answer to the problem.

The solution was to soak the as-produced UO_3, in uranyl nitrate. The resulting product is as reactive as orange oxide produced by the pot process, and it can be processed to metal-grade green salt while using only minimum required amounts of hydrogen fluoride.

This answer to the reactivity problem was found at approximately the same time that the closing of the Weldon Spring facility was announced. When the plant operation was terminated, additional work was still needed on one remaining production problem -- that of finding optimum operating conditions in the plant-scale system.

Conclusions

Comprehensive research activities and pilot-plant work at both Weldon Spring and Destrehan Street resulted in the successful development of an operational, plant-scale fluid-bed denitration system to produce uranium trioxide for subsequent use in the production of uranium metal.

The program's main objective -- to develop a fluid-bed denitration process which would offer advantages making it more practical than the pot process -- was achieved by the Mallinckrodt work. The major advantages of the fluid-bed process, which were established during the extensive development effort in St. Louis and at Weldon Spring, are:

(1) The system is more economical than the pot process. Operating, equipment, and maintenance costs are lower.

(2) The process and equipment provide more control over operating conditions than the pot system offered.

(3) Fewer health and safety precautions are necessary because the closed system reduces possible hazards from dust and fumes.

Production And Costs

Production and cost details are contained in Section IV.

Related Documents

The following numbers refer to bibliography entries which are the principal sources for more detailed information on the fluid-bed-denitration development work described in this subsection: 8, 10, 16, 30, 66, 67, 80, 130, 154, 163, 165, 166, 201, 222, 223, 224, 228.

Additional references may be found in some of the sources indicated above.

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Section II
Part 7

DEVELOPMENT OF THE FIRST FACTORY PROCESS FOR PRODUCING BROWN OXIDE

In addition to developing a commercial ether-extraction process and a practical process for converting uranyl-nitrate into orange oxide, a third important achievement by Mallinckrodt Chemical Works in its initial work on the Manhattan Project was the development of a plant process for producing uranium dioxide (brown oxide, or UO_2).

Background

Until April, 1942, when Mallinckrodt was called upon to provide highly purified uranium compounds for use in the experimental nuclear reactor being developed at the University of Chicago, there had been relatively little work on the reduction of uranium trioxide (orange oxide, or UO_3) to uranium dioxide.

Although the literature contained some information on the preparation of brown oxide by reacting orange oxide with hydrogen as the reducing gas, Mallinckrodt had to evaluate the operation and develop a process which could incorporate the reduction into a practical, factory-production system.

The Basic Process

In the basic process which Mallinckrodt developed, uranium trioxide, which was produced by the pot process, was put into stainless-steel trays in a stainless-steel box. The box was placed in a furnace where, at a relatively high temperature, the reducing gas was passed over the UO_3.

The orange oxide reacted with the gas to produce the sought-after uranium dioxide.

Initial Research Efforts At Mallinckrodt

Because the University of Chicago group needed the uranium products urgently, and because of the consequent pressure toward immediate development of a UO_2-production process, Mallinckrodt's initial research work was concerned primarily with investigating the conditions necessary to achieve an efficient reduction reaction using hydrogen gas as the reducing agent.

In the first experiments, a small stainless-steel tray filled to various depths with orange oxide was used. The primary factors examined were: (1) the optimum operating conditions, including temperature, thickness of the UO_3 layer in each tray, and the heating time required (to attain 95 per cent or better UO_2 assay material); and (2) the extent, if any, of corrosion of the stainless-steel trays.

After a number of experiments, the conditions finally used in the laboratory were: a temperature of 1300°F during a four- to five-hour heating period; cooling to room temperature before opening the box (to prevent oxidation of the dioxide); and a $1\frac{1}{2}$-inch orange oxide layer. These conditions during reduction resulted in the production of a 96-98 per cent UO_2 assay material.

Changes Upon Plant Start-Up

When the plant was started up, it was necessary to increase the temperature to 1500°F for an eight-hour period to obtain satisfactory brown oxide (95 per cent or better UO_2). The trays were filled to a depth of $1\frac{1}{2}$ inches to 2 inches, and the amount of hydrogen required for successful reduction was changed to nearly double the theoretical requirement.

Early in 1943, additional studies were conducted to examine the effects of temperature and nitrate on UO_3 reduction with hydrogen. It was found that the reduction could be accomplished at a temperature considerably lower than 1500°F, and that the reduction was apparently unaffected by different amounts of nitrate present.

The Plant Process

Figure II-7.1 illustrates the type of equipment used in the early plant operation.

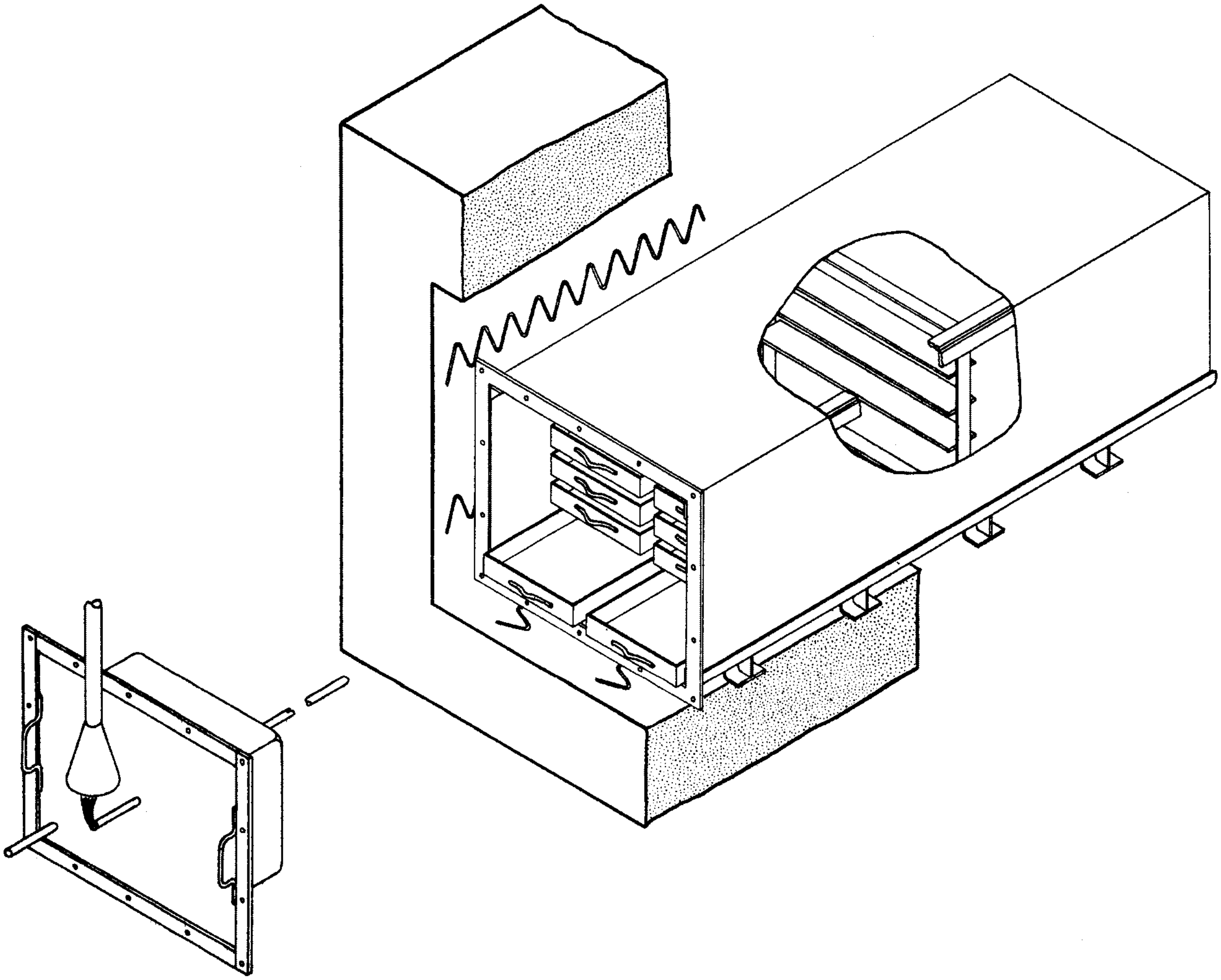

Figure II-7.1 FIRST FACTORY BROWN-OXIDE PRODUCTION employed batch reactors such as this one.

The plant reactor was a stainless-steel box, approximately 13 inches high, 17 inches wide, and 47 inches long. One end of the box was welded shut, and the other was fitted with a bolted end plate containing inlet and exit pipes for the reacting gas.

The orange oxide to be reduced was loaded into eight stainless-steel trays which were placed in a rack inside the reactor box. The box was then closed and put into an electrically heated furnace which was thermostatically controlled.

Residual air in the reactor was driven out by running CO_2 through the box. Then the reducing gas was allowed to enter the reactor and flow over the orange oxide. The unreacted gas was burned continuously as it left the reactor box through the exit pipe.

After a specified heating time, the box was removed from the furnace and cooled both in air and under a water spray. When the box had cooled to the proper temperature, the reacting gas was turned off. Then CO_2 was blown through again to remove the reacting gas remaining in the box. Finally, the brown-oxide product was removed.

Use Of Cracked Ammonia As The Reacting Gas

In the fall of 1942, there was an acute shortage of hydrogen gas, and the possibility of using dissociated (cracked) ammonia to replace hydrogen as the reducing gas was investigated. The dissociated ammonia performed quite well, and an ammonia cracker was installed as part of the system.

At first, little fundamental work was done to determine the best conditions for reduction using the cracked ammonia. However, it became apparent early in 1943 that production demands could not be met under the operating conditions then practiced.

Therefore, a program of experiments was carried out to examine the factors affecting reduction in the furnace and to examine the conditions necessary to meet production requirements.

The results of the investigations showed that with a few changes in operation and by redesigning parts of the reactor box, it would be possible to increase the plant's capacity.

It was concluded that the furnaces should be run on a different time cycle, the weight and distribution of the UO_3 in the box should be changed, and the specified method of operation should be adhered to carefully. Design changes in the equipment were made to improve the gas flow.

After additional experiments, it was decided to change the operation by starting the box at its peak pressure and stopping the gas flow at the end of four hours (two hours before the box was to be removed from the furnace.) The results of these changes were good and reduced the consumption of cracked ammonia.

Investigation Of Other Potential Reducing Gases

During a period of three years, in addition to evaluating the use of hydrogen and cracked ammonia as reducing agents, four other gases were extensively studied to determine their potential values for use in reducing orange oxide to brown oxide. The other gases were anhydrous ammonia gas, wet hydrogen gas, artificial gas and natural gas.

The research was carried out in both the pilot plant and the plant.

The results of the research indicated that cracked ammonia was the most economical, most readily available, and most easily usable of the gases evaluated.

With the use of anhydrous ammonia, a larger quantity of the gas would have been required -- in comparison with cracked ammonia -- to achieve a given assay.

The research indicated that the use of wet hydrogen gas could offer possible advantages as a reducing agent in a reduction unit designed to use a series gas-flow arrangement of boxes.

Artificial and natural gas were found unsatisfactory as reducing agents, because their use would result in the formation of heavy carbon deposits which would flake and contaminate the UO_2 product.

Conclusions

The batch process which Mallinckrodt developed for reducing uranium trioxide to uranium dioxide was simple and effective.

The method, with improvement modifications, was used continuously for seven years, from 1942 until 1949, to produce the UO_2 required for the United States atomic energy activities.

In his official government report, Atomic Energy For Military Purposes, which was completed in 1945, Henry DeWolf Smyth commented on the brown oxide produced by Mallinckrodt. He said, "This oxide is now used as a starting point for all metal production, and no higher degree of purity can be expected on a commercial scale."

In 1948-1950, when successful continuous processes were developed to manufacture green salt, significant advantages were indicated in a potential system which would combine a continuous UO_2 process with a continuous UF_4 process. Thus, work was begun to replace the batch UO_2 process with a continuous method. (See Section II, Part 9.)

Production And Costs

Production and cost details are contained in Section IV.

Related Documents

The following numbers refer to bibliography entries which are the principal sources for more detailed information on the batch process for producing brown oxide: 6, 8, 9, 10, 11, 15, 41, 43, 67, 70, 95, 99, 111, 113, 114, 115, 162, 172, 185, 186, 201, 222, 223, 224.

Additional references may be found in some of the sources indicated above.

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Section II
Part 8

FIRST FACTORY PRODUCTION OF GREEN SALT IN ST. LOUIS

The start of Mallinckrodt Chemical Works' uranium-tetrafluoride (UF_4, or green salt) plant in April, 1943, represented the culmination of a "crash program" by the Company to install a commercial, batch-type green salt operation in St. Louis.

In the years from 1943 to 1951 (when Mallinckrodt introduced the stirred-bed green-salt reactor), the Company improved the process and equipment, and employed the batch technique to produce a major portion of the green salt used for the United States' atomic energy operations.

Background

Early experiments to develop a process for converting highly purified uranium dioxide (UO_2, or brown oxide) to uranium tetrafluoride, which is commonly known as green salt*, were undertaken by researchers at Mallinckrodt, Johns Hopkins University, E.I. du Pont de Nemours & Company, Linde Air Products Company, Harshaw Chemical Company, and others. Wet methods and high-temperature, gas-solid reactions were investigated.

The dry process which was finally selected for commercial production involved the high-temperature reaction of uranium dioxide with anhydrous hydrogen fluoride.

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*An early type of "green salt," potassium uranium pentafluoride (KUF_5) -- not what is referred to as green salt (UF_4) today -- was produced in 1942 by Westinghouse, for use in their metal operations. The process employed the rays of the sun in a photochemical reaction of uranium oxide with potassium fluoride. Because this was, literally, a "fair weather" process, quantity production became a serious problem. By late summer, 1942, the uncertainty of sunlight and the availability of uranium tetrafluoride, which Westinghouse could use instead of KUF_5 in their metal operation, led to the abandonment of the sunlight process.

By late 1942, this process was adopted by Mallinckrodt, du Pont, Harshaw and Linde for large-scale production of UF_4.

The Basic UF_4 Process Employed At Mallinckrodt

In the basic, batch-type, green-salt production process initially adopted by Mallinckrodt, anhydrous hydrofluoric acid was evaporated and passed over heated brown oxide, which was contained on graphite trays in a graphite reactor.

The hydrogen fluoride (HF) gas reacted with the brown oxide to form the sought-after green salt product, uranium tetrafluoride. Water, which was a by-product of the reaction, and excess hydrogen-fluoride gas were neutralized and discarded.

Pilot-Plant Equipment

The primary components of the initial pilot-plant reactor equipment were: a furnace; a graphite box, which was contained in the furnace, and in which the hydrofluorination reaction took place; and graphite trays, which held the brown oxide in the graphite box. A wooden absorption tower was used for neutralizing the waste gases from the reaction.

Furnace

The furnace consisted of a fire-brick shell in which a steel "coffin" was mounted on grate bars. The coffin had a 21-inch square cross section and was approximately 5 feet long. One end of the coffin was fitted with a door, and the other end had two 1½-inch steel pipes welded into it and protruding through the furnace wall. These pipes held carbon tubes which were used to introduce the reactant gas and to remove the waste gases.

Two gas burners were installed under the coffin to provide heat for the reaction. After the first experimental runs, one of the burners was removed, and the remaining one was centered under the coffin.

Temperature in the furnace was controlled manually by regulating the gas and air flow. A thermocouple placed near the bottom of the coffin was used to measure the heat generated by the burner. Heat distribution in other sections of the coffin was determined by a thermocouple mounted in a pipe installation designed to facilitate the measurement of temperatures in various locations throughout the coffin.

Graphite Box And Trays

The graphite box in which the hydrofluorination of the brown oxide occurred was constructed of graphite sheets one inch thick. The box was entirely closed on its outer surfaces.

Four graphite trays were installed within the box to hold the brown oxide charge. The trays were staggered to force the reactant gas to pass over all of the charge material. During operation of the reactor, the reactant gas, which was introduced into the graphite box at one end of the bottom tray, passed over the entire length of the tray, then up to and over the length of the second, third and fourth trays respectively.

After it had reacted with the brown oxide in the fourth tray, the gas, together with the water vapor that formed during the reaction, left the box from the same end at which it had entered. (See Figure II-7.1, Section II, Part 7)

Two types of graphite boxes were used during the pilot-plant experiments. One type employed solid side pieces for supporting the trays. When the reactor was operated, brown oxide and green salt became wedged between the side pieces and the trays. This caused the trays to become jammed and made them difficult to remove. In addition, the expansion of the material that was wedged between the trays and the rough handling required to remove the trays caused some breakage to occur.

To alleviate these problems, an improved box was designed, developed, and tested. In the new box, each tray was supported upon the one below by a narrow graphite plate, the height of which was equal to the distance between the trays. The span of each tray was the same as the width of the box, and each tray's edge formed a portion of the side of the box. This design enabled each tray to be removed easily together with its own two side pieces. There were no cracks or recesses where green salt or brown oxide could accumulate, and cause the tray to become jammed.

Waste Gas System

Waste gases, which were composed of hydrogen fluoride and water vapor, were removed from the furnace and conveyed to a wooden absorption tower. The piping originally installed for this system was made of iron; however, corrosion and scaling of the material caused plugging to occur. To eliminate this problem, brass piping was installed as a replacement for the unsatisfactory iron pipe system.

The wooden absorption tower was approximately one foot square by 15 feet in length. The tower was suspended over and partly within a 300-gallon iron tank which was filled with lime slurry. The lower end of the tower dipped below the liquid level of the slurry.

Slurry, which was drawn off the bottom of the tank, was pumped to the top of the tower where it was applied to neutralize the HF off-gas. The top of the tower was vented to allow the neutralized gas to escape into the atmosphere. After it had been used, the slurry was discarded.

Development Of Effective Reactor Operating Conditions

In addition to designing and developing the equipment in which to carry out the hydrofluorination reaction, the other primary objective of Mallinckrodt's development activities was to establish the most effective procedures for production operation.

Mallinckrodt's laboratory and pilot-plant studies showed that the most important operating variables in the hydrofluorination reaction were: temperature; the quantity of hydrogen fluoride employed during the reaction; the length of time the powder was retained in the reactor; the arrangement of the brown oxide in the reactor; and the degree of reactivity of the brown oxide charge.

Another "Crash Program" To Achieve Plant-Scale Production

Mallinckrodt started plant-scale production of green salt in April, 1943 -- only a few months after the Company had started laboratory work to set up a green salt production process.

The need for green salt to meet the demands of the war effort required the Company to carry out a "crash program" comparable in many respects to the spectacular fifty-day uranium-purification scale-up, from laboratory to ton-per-day production, in the spring of 1942 (see Section II, Part 2). Many out-of-the-ordinary methods were used to put the plant in operation as rapidly as possible. For example, as in the earlier "crash" effort to set up the ether extraction operation, instead of using detailed engineering drawings, equipment-installation plans frequently were sketched roughly on small sheets of paper or were chalk-drawn on the walls and floors of the building that Mallinckrodt leased for construction of the green-salt facilities, the sash and door works adjacent to the Company's main St. Louis plant.

Mallinckrodt engineers and other technical personnel provided around-the-clock supervision and on-site answers to any problems that occurred during the fast-paced project.

Process And Equipment Improvements

The type of batch reactor used for plant production is shown schematically in Figure II-8.1.

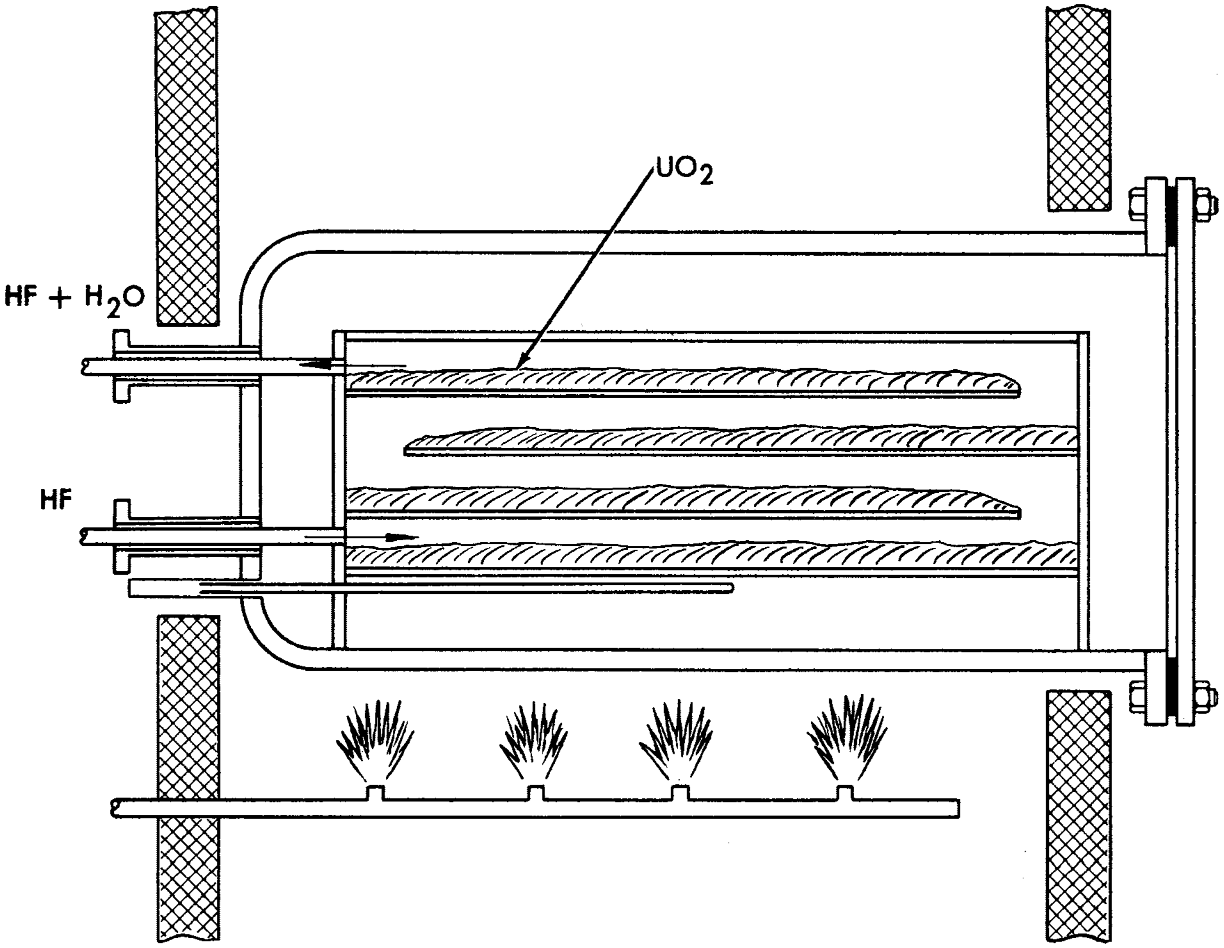

Figure II-8.1 EARLY PRODUCTION OF GREEN SALT was a batch process carried out in assemblies such as the one shown here.

The reactor unit was made of firebox-quality steel. It was approximately 24 inches high by 20 inches wide by 68 inches long. It had a hinged, gasketed door and was mounted in a gas-fired furnace.

At first, graphite tray assemblies, similar to those described above for use in the pilot plant, were placed in the reactor box to hold the brown oxide charge. In 1944, Mallinckrodt replaced the graphite trays with trays made of Monel, a material that was less expensive and less subject to breakage than graphite.

In the original plant, hydrogen gas was fed into each reactor separately through steel pipes in parallel flow. Mallinckrodt improved this process and achieved more economical use of HF by installing a series gas flow to reactor banks consisting of five reactors. Late in 1943, the Company made another improvement to the system by replacing the steel pipes with pipes made of copper. The copper pipes were not only more durable -- and, in this sense, more economical -- but they were also less hazardous than the steel pipes. The hydrofluoric acid corroded the steel pipes, making pin holes. Personnel were in jeopardy of being burned by acid that could come through the pin holes.

In the production operation, brown oxide was supplied to the green-salt plant in drums, each of which contained a 75-pound tray load. The UO_2 was spread uniformly over the reactor trays with a comb-like device which formed a series of ridges on the surface of the brown oxide. This uneven surface exposed a larger area of the charge to direct gas contact than a smooth surface would have exposed.

A controlling thermocouple located below the bottom maintained a steady reactor temperature of $1070^{\circ}F \pm 50^{\circ}F$. Mallinckrodt conducted studies which showed that higher temperatures would have caused excessive caking on the surface of the powder, and incomplete conversion of the brown oxide in the lower part of the bed. Lower temperatures would have required that the brown oxide be retained in the reactor for a longer period to obtain a satisfactory degree of conversion.

At the temperature specified above, and when the hydrogen fluoride gas was to be discarded after passing through five reactors, retention time for the powder was approximately 20 hours. When larger numbers of reactors in series were served by the same gas stream, longer retention times were required. Thus, when 15 reactors were on a single gas stream, each reactor had to be retained in the furnace for approximately 40 hours.

The HF gas fed to a reactor bank was controlled to keep the total supply equal to approximately two to three times the theoretical amount of gas required to convert brown oxide to UF_4. With a bank of 15 reactors in series, the gas supply could be as low as 1.5 times the theoretical amount necessary.

In the original plant, the gas flow to the reactors was controlled by two pressure gauges which measured the drop in pressure across an orifice. In 1947, Mallinckrodt improved the gas-flow control by replacing the orifice meters with Hastelloy C rotameters.

Conclusions

In April, 1943, approximately one year after its successful fifty-day scale-up of uranium purification to produce UO_2 in tonnage quantities for the Manhattan Project, Mallinckrodt completed another "crash program" to install a batch-process, green-salt production plant.

The plant served the nation by producing a major part of the uranium tetrafluoride for atomic energy operations from 1943 until 1951. During that period, the Company improved both the process and the equipment to make the batch green-salt operation more economical and safer.

In 1951, the Company replaced its batch-type green-salt reactors with new, continuous-process, stirred-bed, screw-conveyor green-salt reactors which Mallinckrodt had invented, designed and developed.

Production And Costs

Production and cost details are contained in Section IV.

Related Documents

The following numbers refer to bibliography entries which are the principal sources for more detailed information on the batch, green-salt production process discussed in this subsection: 8, 10, 11, 67, 90, 98, 110, 112, 116, 117, 134, 138, 185, 186, 201, 216, 222, 223, 224.

Additional references may be found in some of the sources indicated above.

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Section II
Part 9

DEVELOPMENT OF THE CONTINUOUS-PROCESS, STIRRED-BED REACTOR FOR THE PRODUCTION OF GREEN SALT AND BROWN OXIDE

In 1948, Mallinckrodt Chemical Works began experimental work in search for a continuous process for the production of uranium tetrafluoride (UF_4, or green salt). The work ultimately resulted in the development of a unique, stirred-bed reactor which employed a screw-conveyor for transporting the materials in process.

In the course of the work, the equipment and the process were adapted to the production of uranium dioxide (UO_2, or brown oxide). By combining the continuous UO_2 process with the continuous UF_4 process, Mallinckrodt was able to build a plant in which UF_4 was produced continuously using uranium trioxide (UO_3, or orange oxide) as the starting material.

Background

In 1943, Mallinckrodt first installed batch reactors for the reaction of brown oxide with anhydrous hydrogen-fluoride (AHF) gas to produce green salt. (A more detailed discussion of the batch green-salt process is presented in Section II, Part 8. The process worked well, but by 1948, increased oxide production threatened to exceed the capacities of both the green salt and the metal plants.

Faced with a need for greater production capacity, Mallinckrodt technical personnel proposed the idea of stirring the oxides and green salt, while conveying them countercurrent to the reacting gases. From 1948 until 1951, through its laboratory and pilot plant work, the Company developed the new concept into what was considered by authorities to be a "radically novel" green salt process -- the stirred-bed, screw-conveyor reactor process.

The Basic Stirred-Bed, Screw-Conveyor Reactor Concept

The basic stirred-bed reactor concept that Mallinckrodt developed for green-salt production involved mechanically conveying UO_2 through a heated tube at a specific rate by means of a screw/agitator mechanism, and at the same time, passing anhydrous hydrofluoride (AHF) gas through the tube in a countercurrent manner. A typical stirred-bed reactor employing this process is shown in Figure II-9.1. The continuous stirred-bed, screw-conveyor reactor for brown oxide production is nearly identical to the green-salt reactor.

The type of rotating, stirred-bed agitator/conveyor located inside the reactor tube is illustrated in Figure II-9.2.

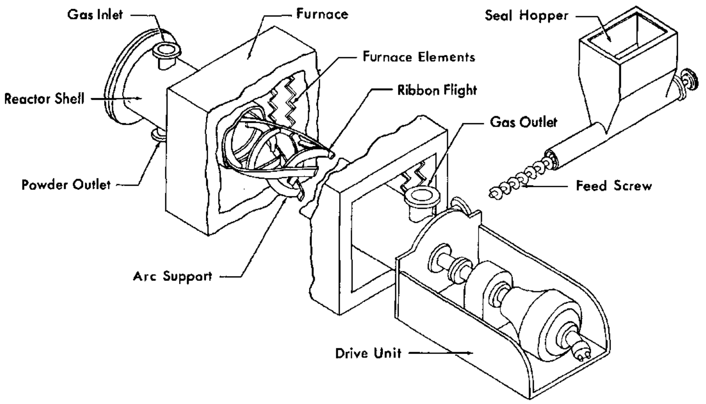

Figure II-9.1 TYPICAL STIRRED-BED GREEN-SALT REACTOR ASSEMBLY.

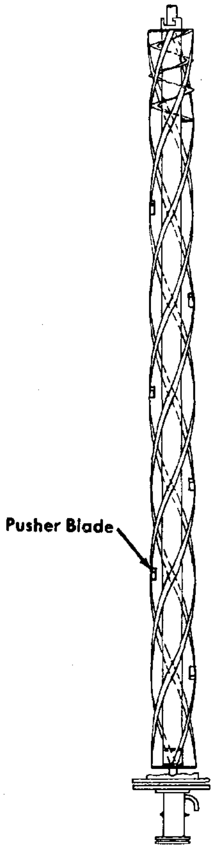

Figure II-9.2 AN AGITATOR/CONVEYOR of the type shown here rotates inside the stirred-bed reactor tube.

In all of the development work, the AHF gas was introduced into the reactor at the product-discharge end (to give the desired countercurrent flow), and the exhaust gases were removed at the UO_2-feed end of the reactor. Heat was applied externally to the reactor during the process.

Objectives Of The Research And Development

In all of the work that Mallinckrodt did to develop the continuous green salt method, the chief objectives were: (1) to determine whether the continuous process would be practical; and (2) if the process were practical, to develop the operating procedures and process-equipment designs on which to base construction of a large-scale production unit.

The construction of Plant 7 in 1951 marked the realization of these goals.

Extensive Pilot-Plant Develpment

The extent of the pilot-plant development is illustrated by the fact that a total of 36 different conveyors and four different basic reactor units (with additional variations) were designed, fabricated, tested and evaluated.

The Conveyor

During the development program, the conveyor design was found to be an exceptionally critical variable in the operation of the stirred-bed reactor. Therefore, the greatest part of the pilot-plant work was devoted to developing a suitable conveyor.

An ordinary screw·reactor would have been totally unsatisfactory because it seemed to cause or allow caking under almost any conditions, and did not provide very good agitation.

It was finally determined that the conveyor to be used in the continuous reactor should have the following characteristics:

- To achieve complete hydrofluorination, it must be able to convey material slowly through the reactor.
- To produce a uniformly hydrofluorinated product, the conveyor must provide good mixing.
- It must be able to revolve at a speed high enough to prevent caking of the UO_2 feed and the UF_4 product.

- To enable the hydrogen fluoride to come into intimate contact with the brown oxide, the conveyor should allow the reacting gas an unrestricted flow.

- The conveyor must be made of an alloy that has: a high creep strength at elevated temperatures, reasonable workability, and resistance to AHF and HF-H_2O at high temperatures.

A total of 36 different conveyors were tried in the pilot plant.*

The final four conveyor designs studied were of the compound-conveyor type. Each of them had three flights with a 6-foot, 9-inch pitch, and each was about 11½ inches in diameter. The Number 36 conveyor, which was the last conveyor made for the pilot plant and which was subsequently operated as a production unit, had twelve 1¼-inch by 4-inch by ½-inch deflectors, which were welded to the inside surfaces of the flights, to help convey and mix the reactor materials. The deflectors were spaced equidistant between the feed and the discharge inner ribbons. "Y" braces were installed about every 14 inches for strength and support.

The Reactor

During the reactor-unit development program, variations in design factors that were investigated included: materials of construction, the size of the inside diameter of the reactor tube, the length of the reactor, the length of the heated section, the methods of heating the reactor and the arrangement of the heating units, instrumentation devices for controlling and monitoring temperatures, and AHF flow. Mechanical factors examined included: methods for feeding UO_2 and AHF into the reactor, methods for driving the conveyors and for connecting them inside the reactor, and methods for removing and handling the discharged product and exhaust gases.

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*Descriptive detail on each of the 36 conveyors which were investigated in the pilot plant and comments on the results they gave may be found in USAEC report NYO-1316, Mallinckrodt Chemical Works, St. Louis, Missouri, December 22, 1950. For more specific data concerning the ongoing development of the continuous process, refer to the series of Mallinckrodt Chemical Works (MCW) reports entitled "Progress Report of Pilot Plant Work on UF_4 Production By Continuous Methods."

The final pilot-plant unit was a double pass reactor. During testing, it produced a large quantity of high quality UF_4. It was later used as a production unit for Plant 4 and as a re-run unit in Plant 7.

Material Of Construction

Selection of a suitable material of construction was one of the chief problems that had to be solved in the development of the continuous-process, stirred-bed reactor. The material selected had to have: extremely low corrosion rates under the reaction conditions; good strength at temperatures ranging as high as 1500°F, and good resistance to a great deal of abrasive action.

During the development program, more than ten different metals were tested as possible materials of construction for the reactor and/or conveyor. The metals tried included: steel for a reactor and conveyor, Hastelloy C for a reactor and conveyor, Monel for a reactor liner and conveyor, aluminum for a reactor liner, manganese plated steel for a reactor, Inconel for a reactor and conveyor, Copel for a reactor and conveyor, Super Nickel (70 per cent copper, 30 per cent nickel) for a conveyor, calorized steel for a conveyor, and Illium R for a reactor and conveyor.

After the long testing program, both Illium R and Hastelloy C were found to be satisfactory for the agitator/conveyor, and Illium R and Inconel were found to be acceptable for the reactor tube.

Temperature Control

Another significant operating variable that was extensively investigated during the development program was temperature control. The temperatures of the reactors influenced both the quality of the product, and the extent of any possible caking and/or plugging difficulties.

Early laboratory work indicated that at 1200°F a 97 per cent conversion of UO_2 to UF_4 could be obtained in less than one-tenth the time required at 750°F. During the early development work, however, attempts to operate at higher temperatures resulted in excessive caking. The caking either stopped the conveyor or resulted in the formation of hard lumps with large sections of unconverted brown oxide in their centers. Difficulties were also encountered in experiments at lower temperatures. The low temperatures resulted in conversion rates so low that, if applied on a plant scale, they would have required unreasonably large amounts of equipment.

Ultimately, the temperature problem was solved by using a graduated temperature ranging from approximately 700°F at the point where the UO_2 was fed to about 1100°F where the UF_4 product was discharged.

Investigations indicated that the basic problem in the conversion of brown oxide to green salt concerned transferring heat away from each particle of UO_2 as it reacts with the hydrogen fluoride.

Because the reaction is highly exothermic, extremely high particle temperatures would have resulted if the heat were not removed at a rapid rate from the vicinity of a reacting particle. The subsequent fusing, which apparently results from the high particle temperature, interfered with the diffusion of gas required to continue the reaction. If many particles reached a high enough temperature, large masses of the material would ultimately fuse and eventually cripple the agitator/conveyor mechanism.

It became evident, therefore, that controlled heating was very important, and for this reason, multi-zone electrical heating was selected for use in the plant design.

The Plant Process

The process and equipment designs that evolved from the pilot-plant development work were reduced to practice and first operated at Plant 7 in 1951. The type of stirred-bed hydrofluorination reactor employed is illustrated schematically in Figure II-9.3.

The type of green-salt manufacturing system which was installed at the Weldon Spring plant is illustrated in the schematic representation in Figure II-9.4. The hydrofluorination reactors were arranged in banks consisting of three horizontal tubes, each of which was 22 feet long by 16 inches in diameter. The reactor bank was arranged in a vertical stack.

The design of the first stirred-bed reactor plant was based on a production of 233 pounds of green salt per hour per reactor bank. This rate was achieved almost immediately upon plant start-up, and as additional experience was gained with the system, production rates increased.

Two major shake-down problems arose in connection with the increase of UO_2 feed rates beyond those specified for the original plant design.

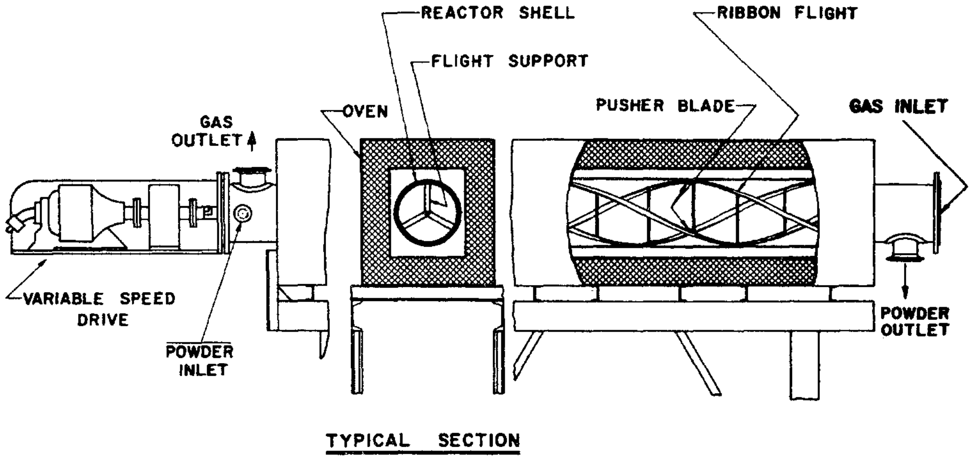

Figure II-9.3 STIRRED-BED PRODUCTION REACTOR FOR PRODUCING UF_4.

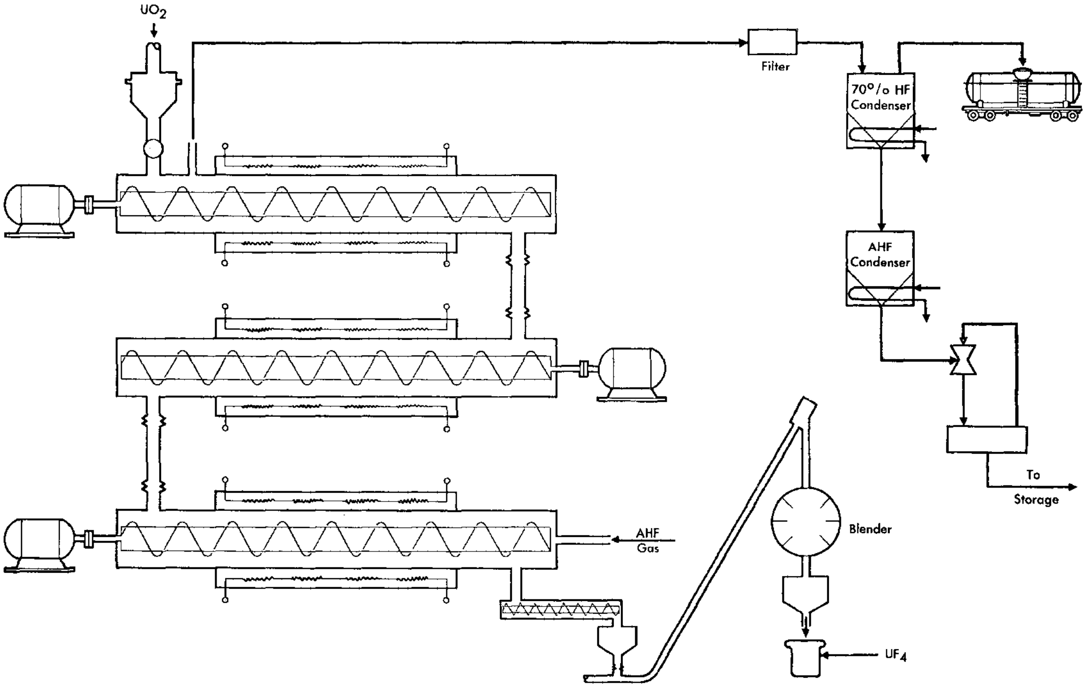

Figure II-9.4 CONTINUOUS STIRRED-BED GREEN-SALT MANUFACTURING SYSTEM.

One of the problems involved low conversion and/or caking, which resulted from difficulty in maintaining a low enough temperature in the first two zones of the first reactor. The second problem involved the return of dust from the off-gas stream to the reactor system. When the carbon-tube filter, which was located above the top reactor, was back-blown, a large quantity of material would suddenly drop into the top reactor tube and place an additional load on that tube's conveyor. Both problems were alleviated through minor changes in the design.

Use Of The Stirred-Bed Reactor For Producing Brown Oxide

Late in 1948, Mallinckrodt attempted to reduce orange oxide to brown oxide in the continuous, stirred-bed, screw-conveyor equipment that was being used in the pilot-plant development of the continuous green-salt process.

Two of the principal objectives of the intital experimental work were: (1) to determine whether low temperature brown oxide could be made in a continuous, screw-conveyor reactor; and (2) to find the most economical hydrogen rate, retention time and reaction temperature for the production of low-temperature brown oxide.

In the technical report which summarized the first experiments, it was concluded that brown oxide could be made in the stirred-bed reactor and that it would "equal or surpass, in UO_2 assay," that which was obtained from the batch equipment.

The plant equipment and procedure that ultimately evolved was similar to that used for the green salt operation.

Conclusions

In conceiving the idea of the stirred-bed reactor, and in subsequently developing the concept and reducing it to practice for the continuous production of both brown oxide and green salt, Mallinckrodt achieved a major breakthrough in uranium production technology.

The advantages of the continuous system were numerous. The brown oxide and the green salt were purer and more uniform in quality than the UO_2 and UF_4 produced by the earlier batch reactors. Equally significant was the fact that the process made it possible for costs to be greatly reduced. Furthermore, with the stirred-bed reactors, the entire production operation -- from UO_3 to green salt -- was safer than ever before.

The process was so successful that it was installed at all subsequent United States green-salt production plants. The equipment is still the industry standard for the hydrofluorination step. The use of fluidized beds for the UO_2 step was started in 1956, and by 1960 fluid beds had replaced screw reactors for brown-oxide production.

Production And Costs

Production and cost details are contained in Section IV.

Related Documents

The following numbers refer to bibliography entries which are the principal sources for more detailed information on the continuous, stirred-bed, screw-conveyor reactor process described in this subsection: 4, 8, 10, 13, 16, 19, 31, 41, 43, 67, 78, 132, 153, 159, 160, 171, 173, 179, 180, 185, 186, 190, 201, 222, 223, 224.

Additional references may be found in some of the sources indicated above.

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Section II
Part 10

ADVANCES IN CONTINUOUS-PROCESS PRODUCTION OF UF_4

DEVELOPMENT OF THE INTEGRATED FLUID-BED SYSTEM

From 1957 to 1963, Mallinckrodt Chemical Works' Uranium Division carried out research, pilot-plant work and plant-scale activities through which the Company developed an operable, integrated system for producing uranium tetrafluoride (UF_4, or green salt) on a continuous basis using pure uranyl nitrate as the starting material. The integrated system combined three separate fluid-bed processes developed by Mallinckrodt -- denitration of uranyl nitrate to produce uranium trioxide (UO_3, or orange oxide), reduction of UO_3 to uranium dioxide (UO_2, or brown oxide), and hydrofluorination of UO_2 to produce green salt -- so that the product of each step was an optimum feed to the succeeding step.

Background

Investigation of the possibility of employing a fluid-bed technique in uranium processing was first started in 1953 at Argonne National Laboratory for use in continuous-process denitration. In addition, a small-scale fluid-bed reactor was designed and used at Argonne for experimentation on the production of brown oxide from orange oxide and on the production of green salt from brown oxide. In 1956 at the Oak Ridge Gaseous Diffusion Plant, Union Carbide Nuclear Company carried out an experimental program which resulted in the successful plant-scale operation of a fluidized-bed reduction reactor designed to use pot-process orange oxide as the feed material.

While Argonne and Union Carbide were studying the feasibility of the fluid-bed concept as an approach to continuous processing, Mallinckrodt already was using a successful continuous process -- the stirred-bed reactor -- which the Company's personnel had conceived and developed during the period from 1948 to 1951. Mallinckrodt used the stirred-bed reactor process for reduction until 1957 and for hydrofluorination until the Weldon Spring plant was closed in 1966. (Details concerning the development of the stirred-bed reactor are presented in Section II, Part 9.)

Denitration of uranyl nitrate to UO_3 remained a batch process. However, the promising investigations at Argonne and at Union Carbide encouraged extensive development by Mallinckrodt to make the entire UO_3-UF_4 production sequence a continuous process at the St. Louis-area uranium processing facilities.

The installation of a pilot-plant fluid-bed denitrator at Mallinckrodt's Destrehan Street plant in 1957 was the first step in the Company's long-term efforts to develop an "integrated" fluid-bed system for producing green salt.

Development Of The Fluid-Bed Denitrator

The nine years of work, from 1957 to 1966, which Mallinckrodt devoted to developing an operable, plant-scale, fluid-bed denitration process represented the largest and most significant part of the Company's development effort on the entire integrated fluid-bed system. A detailed description of Mallinckrodt's extensive program to develop the fluid-bed denitration process is presented in Section II, Part 6.

Pilot-Plant Development Of The Fluid-Bed Reduction Reactor

In 1958, shortly after initial operation of the Destrehan Street pilot-plant fluid-bed denitrator proved the feasibility of the fluid-bed concept, Mallinckrodt started design and construction work on a pilot-plant fluid-bed reactor for reducing UO_3, produced in the fluid-bed denitrator, to UO_2. Although there had been previous work on fluid-bed reduction of pot-process UO_3, there had been no previous development of a fluidized-bed reactor for reducing fluid-bed-produced UO_3.

During the reduction-reactor development program, the effects of altering certain variables were examined to determine which operating conditions would achieve optimum productivity as well as good product assay. The variables investigated included: solids feed rate; bed height and, therefore, active volume of the reactor; fluidizing gas velocity, and the resulting degree of solids mixing; and temperature.

The First Pilot-Plant Reduction Reactor

The original design of the fluid-bed reduction reactor was based on data obtained from Union Carbide Nuclear Company and preliminary laboratory studies or orange oxide produced by the fluid-bed process. A sketch of the pilot-plant fluid-bed reduction system as it was initially designed and built is shown in Figure II-10.1.

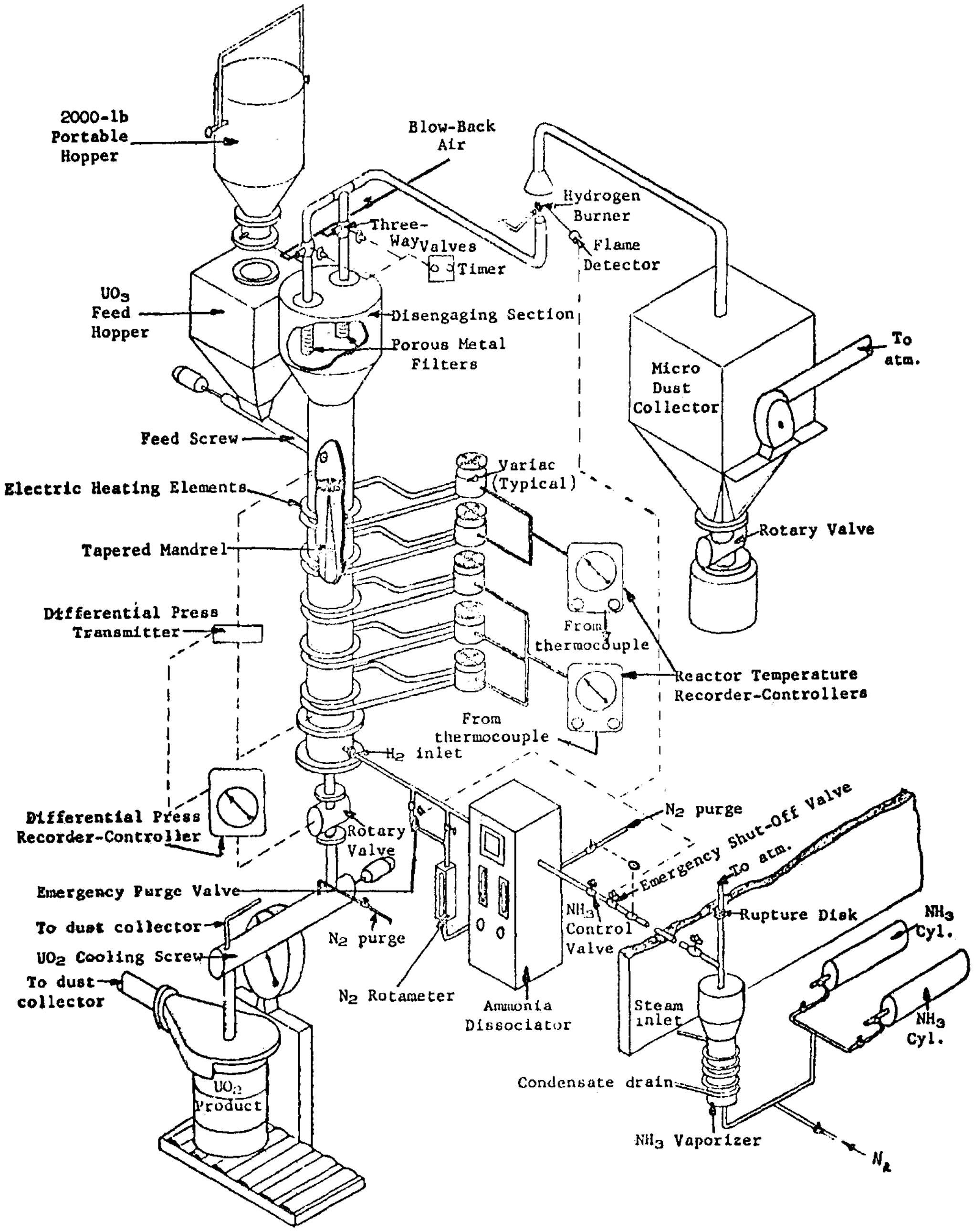

Figure II-10.1 INITIAL DESIGN OF PILOT-PLANT FLUID-BED REDUCTION SYSTEM.

The reactor unit was a stainless-steel pipe 5 feet, 9 inches long, and 5 inches in diameter. Centered within the pipe was a tapered mandrel 53 inches long. The mandrel's bottom diameter was 2.89 inches, and it tapered to a top diameter of 0.63 inch.

The tapered mandrel was a unique feature of the pilot-plant reactor. It was incorporated into the design to control fluidization velocity, and thus, to help prevent turbulent solids mixing which could cause particles of the orange-oxide feed to "short-circuit" to the product line before they had been completely reduced.

During operation, dissociated ammonia was metered into the bottom of the reactor through a gas-distribution plate made of porous metal. Orange oxide was fed to the top of the bed through a 2-inch-diameter feed screw. The brown-oxide product was removed from the bottom of the reactor through three 1-inch-diameter holes which slanted towards a downcomer welded to the bottom of the mandrel. From the downcomer, the UO_2 powder passed through an expansion bellows and a rotary valve to a cooling screw and a drumming station.

A screw-type sampler was located at the cooling screw discharge. Off gases left the top of the reactor through a disengaging section which contained two porous metal filters for removing powder from the off-gases. Electrical heating elements clamped to the reactor shell supplied heat to the reactor at five heating zones.

The Research And Development Program

Much of the initial experimentation was devoted to establishing the operating and design conditions necessary to achieve increased product quality through decreased mixing. A technical report dealing with this aspect of the development effort showed that the single-stage, pilot-plant reduction reactor could be operated to produce a product equivalent to that produced by 13 well-mixed stages in series.

Following the mixing experiments, development activities turned to production of UO_2 as feed for the hydrofluorination reactor. Data derived from this phase of the pilot-plant effort established more firmly the reliability of the system.

As development of the integrated fluid-bed system progressed, it was found that the capacity of the fluid-bed hydrofluorination system exceeded reduction capacity. A larger fluid-bed reduction unit was installed to overcome this limitation. A schematic diagram of the larger reactor is shown in Figure II-10.2.

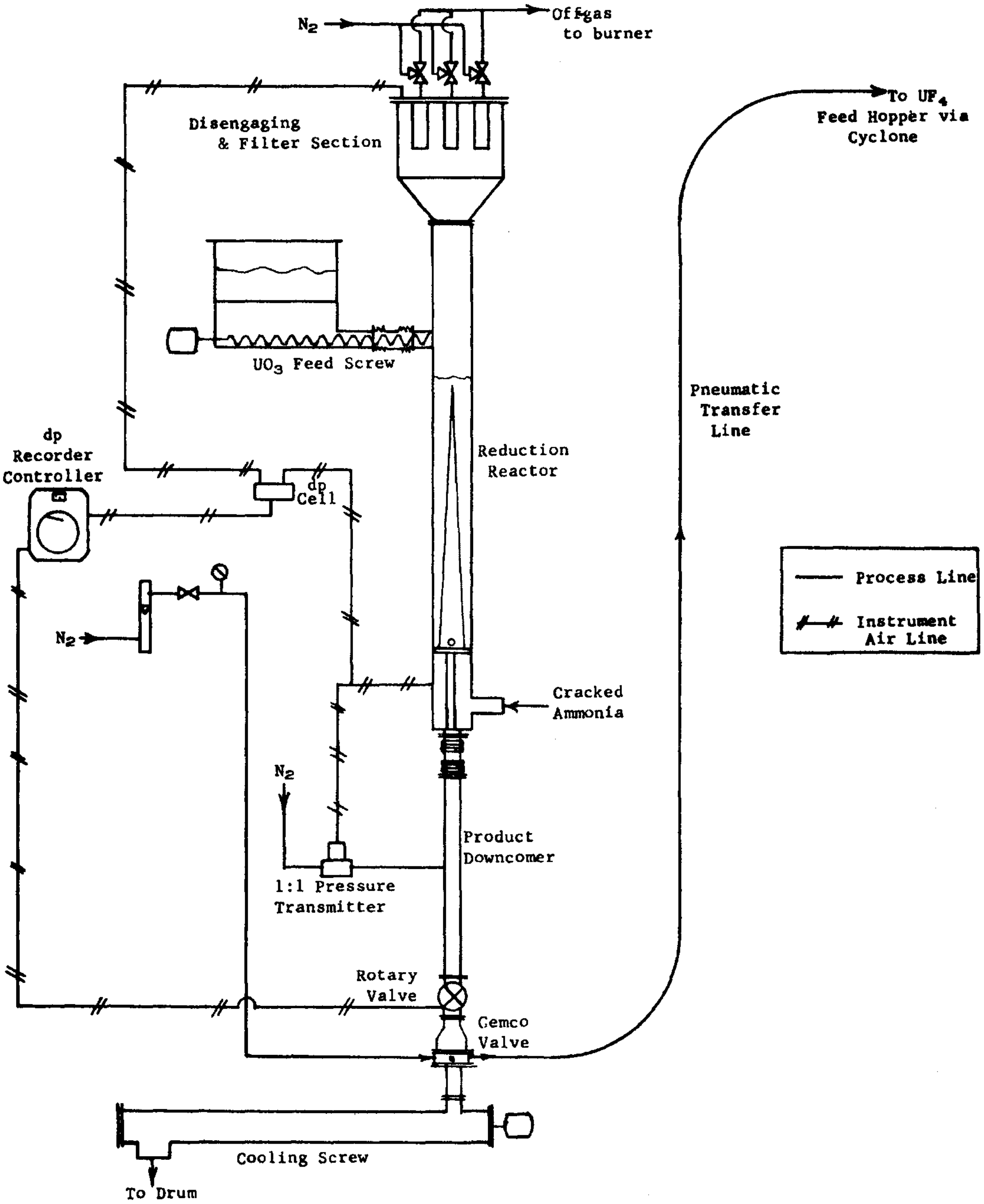

Figure II-10.2 THE MODIFIED PILOT-PLANT FLUID-BED REDUCTION REACTOR was larger than the one in the initial system design shown in Figure II-10.1.

The new unit contained no mandrel, but was built with a removable baffle plate down the center of the reactor tube to a point $4\frac{1}{2}$ inches above the distribution plate. Orange oxide was fed into the reactor by a screw conveyor located 4 feet, 9 inches above the gas distributor. The brown-oxide product overflowed through a downcomer located 4 feet above the distribution plate.

The center-baffle design served to separate the feed and the product, and thus, prevented short-circuiting of unreacted material. Another object of this design was to evaluate the potential of operating the system as two well-mixed reactors in series. The results of mixing studies, however, indicated that under operational conditions, only one-stage operation was actually achieved. After some additional investigation, the two-stage concept was abandoned.

Subsequently, the demands for high-assay UO_2 for hydrofluorination led to a return to the original reactor design. Both the mandrel and the bottom withdrawal system were employed again. Because the size of the reactor shell had been increased, the UO_2 production rate was adequate for the capacity of the hydrofluorination system.

The fluid-bed reduction system that ultimately evolved from the pilot-plant development consistently produced high-quality UO_2. In the final system design, the product was transferred directly to the hydrofluorination system.

The Plant Fluid-Bed Reduction System At Weldon Spring

The first plant-scale fluid-bed reduction system for the St. Louis-area operation was installed in 1957 as part of the construction of the Weldon Spring facility.

When the Weldon Spring plant was built, the plant-scale fluid-bed denitration system had not been fully developed. Therefore, design of the fluid-bed reduction equipment for the plant had to be adapted to operate on pot-process UO_3 feed instead of fluid-bed UO_3. For successful operation with pot-process UO_3, it was necessary to modify the single-stage reduction concept that had been developed in the pilot plant, to a two-stage configuration. A schematic diagram of the production unit is shown in Figure II-10.3.

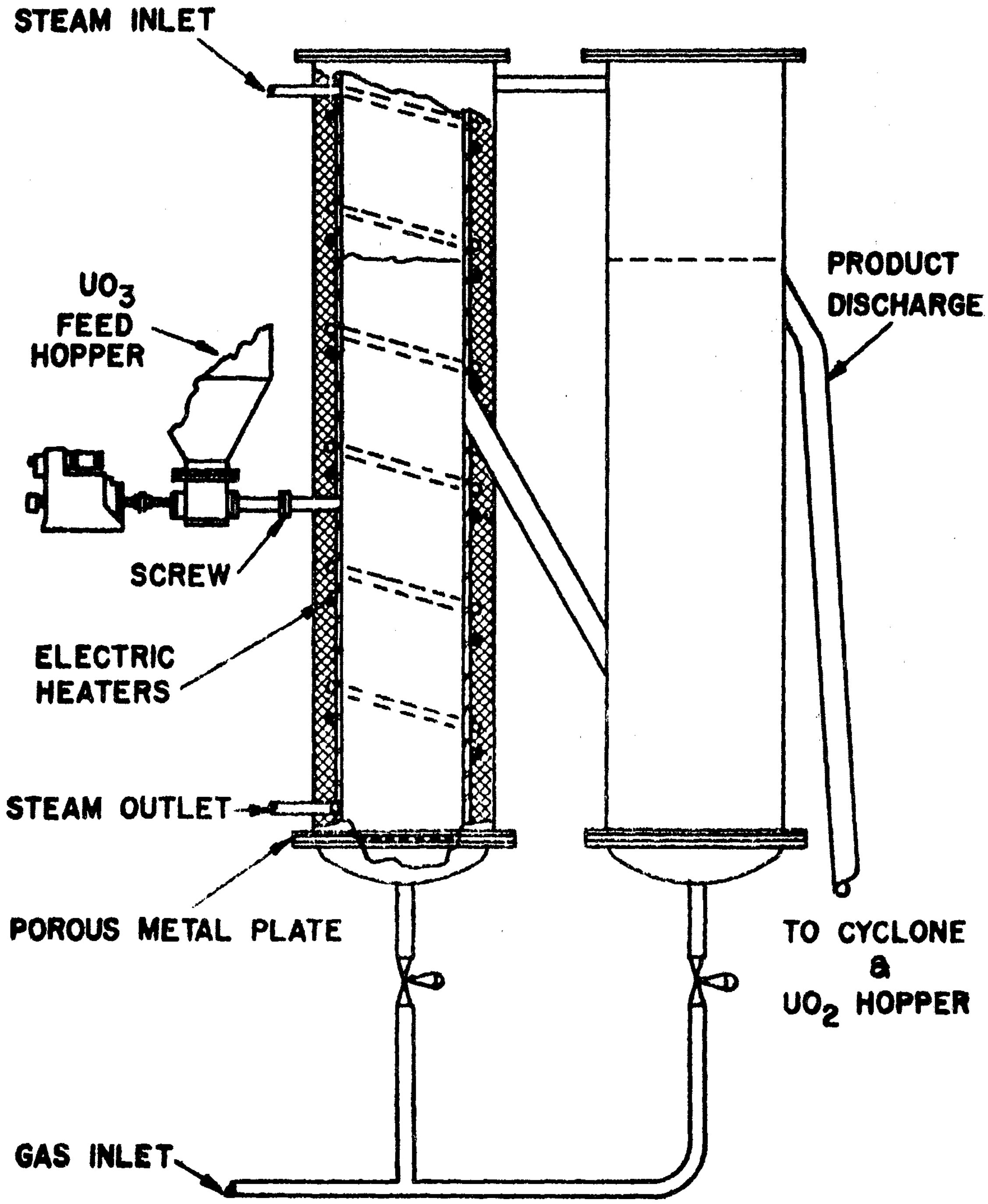

Figure II-10.3 THE FLUID-BED URANIUM-DIOXIDE PRODUCTION REACTOR designed for the Weldon Spring plant was adapted to operate on pot-process UO_3 by employing a two-stage configuration.

The reduction equipment consisted of two vertical stainless-steel cylinders. Each was 6.5 feet high by approximately one square foot in cross-sectional area. Orange-oxide feed entered the side of the first tube where it was partially reacted. The material then overflowed into the second tube where the reaction was completed.

Pilot-Plant Development Of The Fluid-Bed Hydrofluorinator

Mallinckrodt started pilot-plant work aimed at developing an operable fluidized-bed hydrofluorinator, the third major component of the integrated fluid-bed system.

In addition to establishing the physical configuration of the reactor system, the development work was designed to find the specific operating conditions required to produce metal-grade green salt from fluid-bed-produced UO_2. A major part of the investigation focused on experiments involving reactor volume, solids mixing, temperature, velocity of the fluidizing gas, and solids feed rate.

The Pilot-Plant Single-Stage Fluid-Bed Hydrofluorinator

The original hydrofluorinator tested in the pilot plant was a single-stage unit. A diagram of the reactor is shown in Figure II-10.4. As in the fluid-bed reduction unit, a tapered mandrel was incorporated within the reactor tube.

A schematic drawing of the hydrofluorination system as originally built is shown in Figure II-10.5.

During operation, vaporized anhydrous hydrofluoric acid (AHF) was heated to approximately 220°F before metering. The fluidizing gas was preheated electrically in a section of pipe, and entered the bed through a porous metal distribution plate.

A screw conveyor was used to feed uranium dioxide into the reactor. The green-salt product was removed from the bed to a downcomer through four 1-inch holes in the bottom of the mandrel. From the downcomer, the powder entered an expansion bellows and then moved through a rotary valve to a cooling screw which conveyed the green salt to a drumming station.

The reactor was divided into six heating zones, two of which were wrapped with electric heaters, and four of which were contained within a radiant furnace. Thermocouples were used to control the zones.

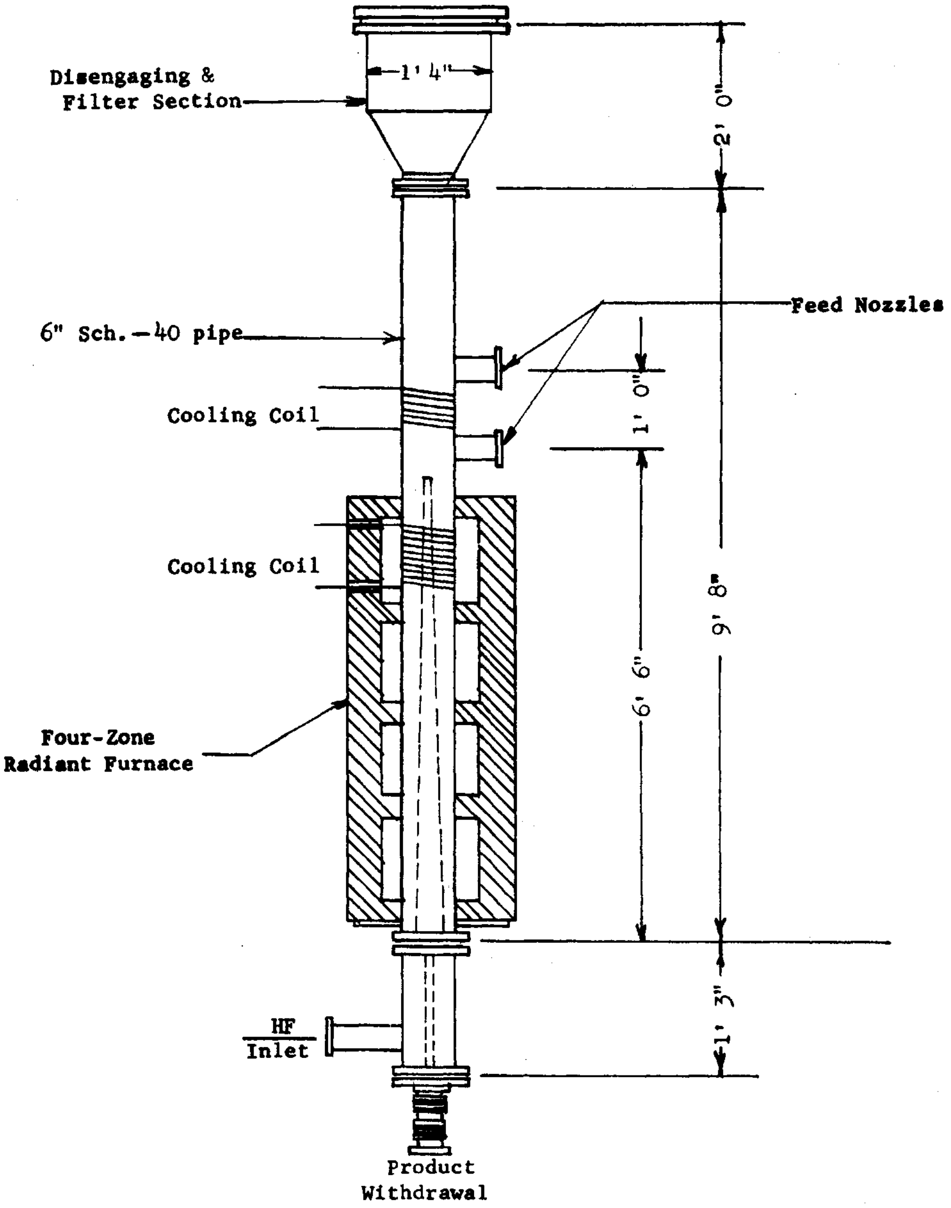

Figure II-10.4 PILOT-PLANT SINGLE-STAGE FLUID-BED HYDROFLUORINATOR.

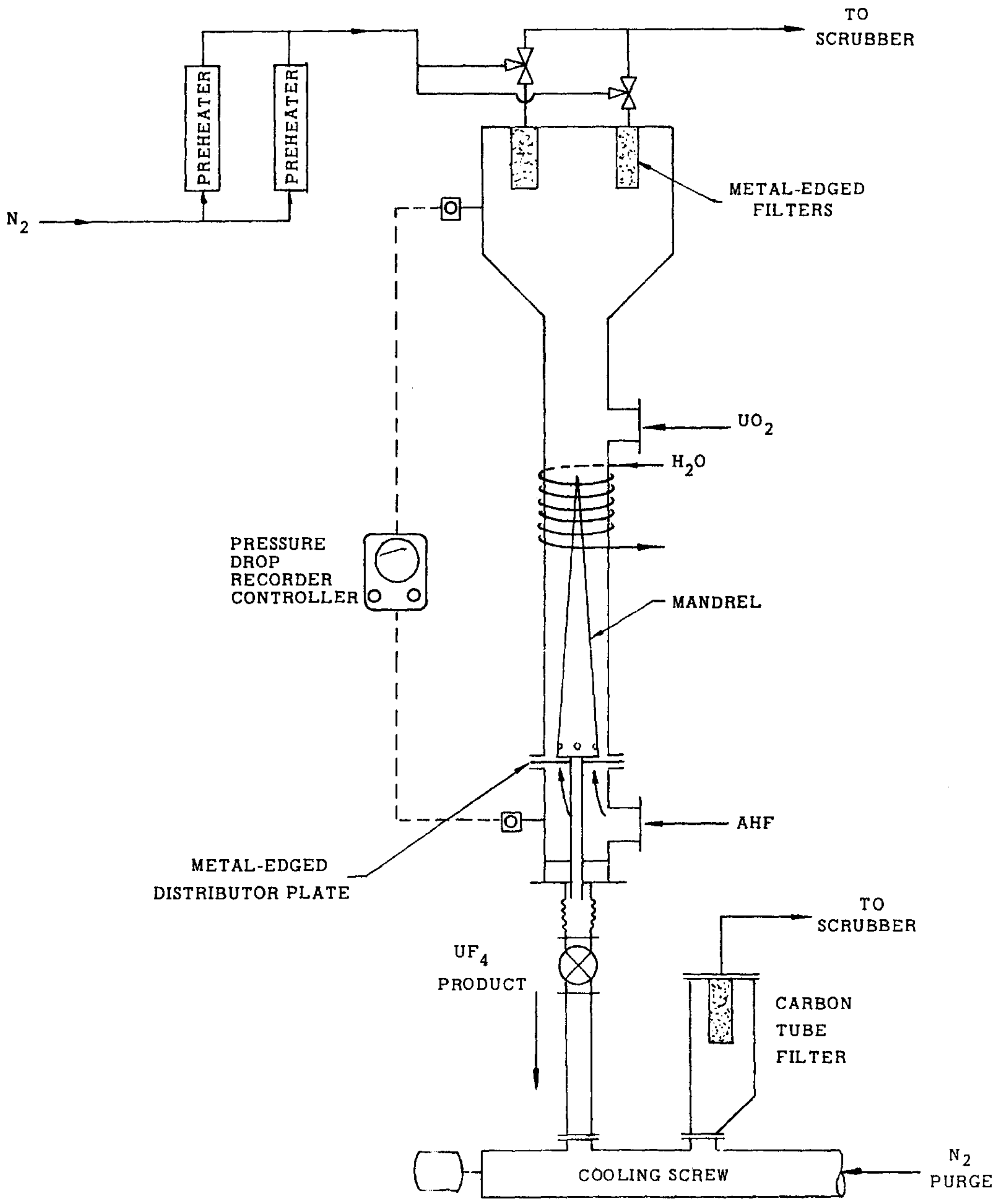

Figure II-10.5 PILOT-PLANT SINGLE-STAGE FLUID-BED HYDROFLUORINATION REACTOR SYSTEM AS ORIGINALLY BUILT.

Off gases were filtered through porous metal filters to remove entrained solids. The off-gas filters were periodically back-blown with nitrogen which was preheated in two parallel electric heaters.

In the initial experiments, operating instability and below-specification product were encountered. After the equipment was modified as shown in Figure II-10.6, the reliability of the operations was considerably improved, but high-quality green salt still could not be produced.

One of the major operating difficulties experienced with the single-stage hydrofluorinator was reactor plugging. Investigation showed that the plugging was caused by build-up of cake on the reactor wall. The caking apparently occurred at any point of high UO_2 concentration.

Because of the plugging tendency, low fluidizing velocities, which retard solids mixing, could not be employed. Instead, high gas velocities were required to achieve the degree of agitation needed to reduce the plugging. With the resulting high degree of solids mixing, acceptable green salt could not be produced in the single-stage reactor.

Development Of The Two-Stage Hydrofluorination Reactor System

Subsequently, a two-stage fluid-bed hydrofluorination system was developed for use in the pilot plant. A schematic drawing of the system is shown in Figure II-10.7.

In the two-stage hydrofluorination design, a new hydrofluorination unit was developed for operation, in series, ahead of the fluid-bed hydrofluorination unit previously used in the single stage system. To minimize powder-caking tendencies, the first stage was designed to operate with the solids in a well-mixed condition. The design of the new reactor unit incorporated internal heat-transfer surfaces to remove more efficiently the heat generated during the reaction.

The initial operation of the two-stage system was completely successful. The equipment performed perfectly, and it produced high-quality green salt.

Additional experimental studies showed that particle size of the UO_2 feed had a significant effect on product quality. Coarser brown oxide with higher incipient fluidizing velocity was found to yield a better green-salt product than fine material.

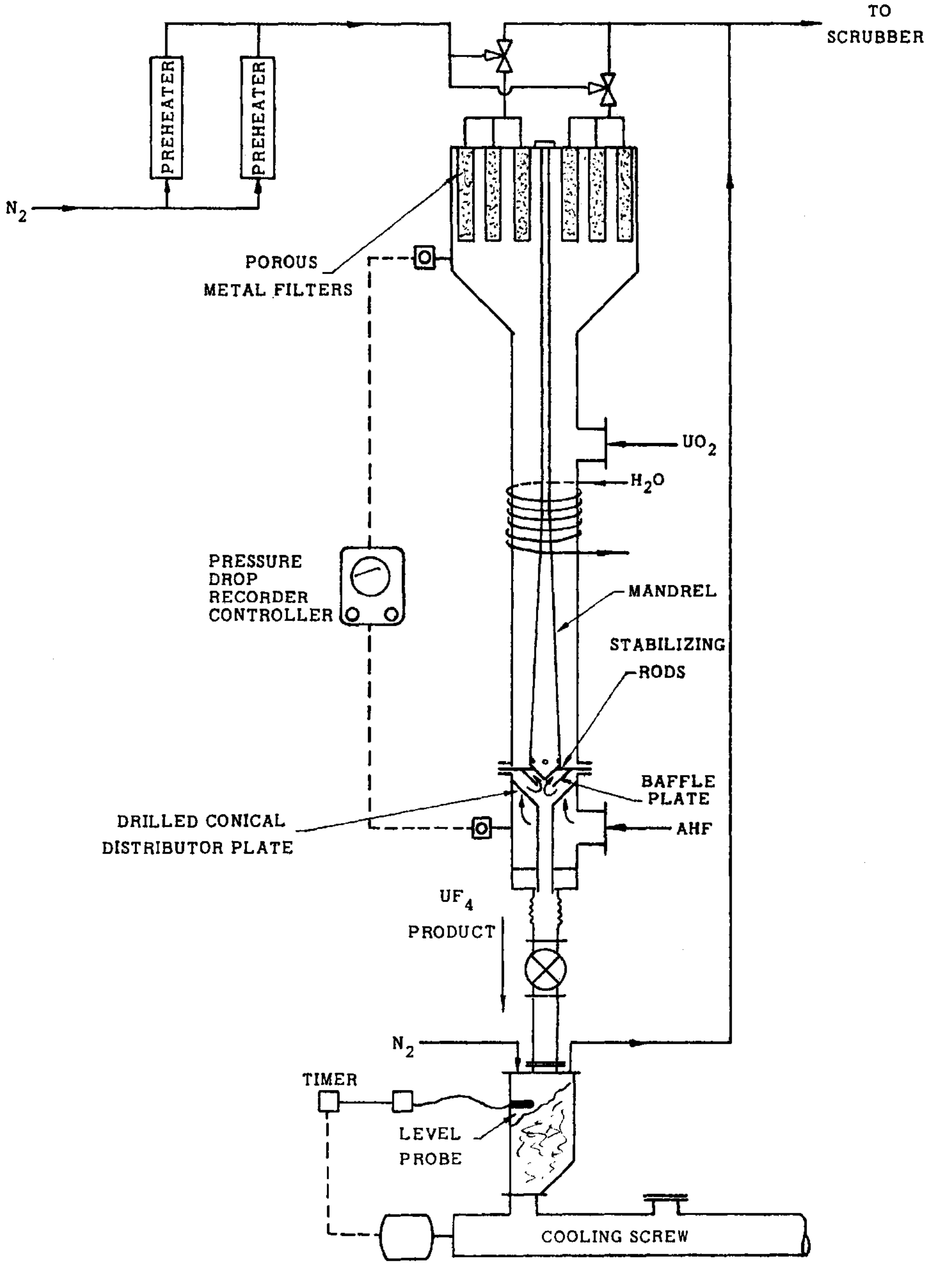

Figure II-10.6 THE MODIFIED PILOT-PLANT SINGLE-STAGE FLUID-BED HYDROFLUORINATION SYSTEM was more reliable than the original system shown in Figure II-10.5.

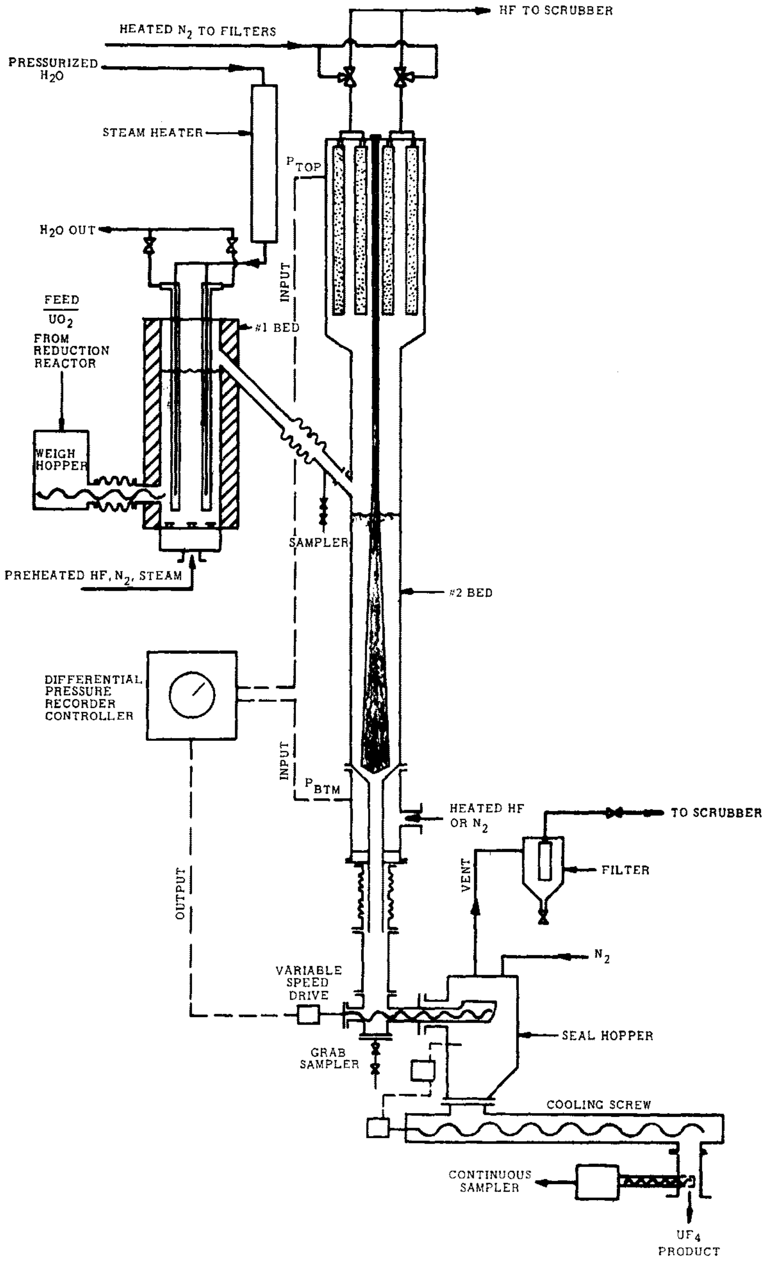

Figure II-10.7 PILOT-PLANT TWO-STAGE FLUID-BED HYDROFLUORINATION SYSTEM produced high-quality green salt.

Operating temperatures were another significant factor which appeared to affect the quality of the UF_4 product. The temperatures used for the reduction process also were found to be important. Experiments indicated that the optimum reduction temperatures for subsequent production of specification UF_4 were in the range of 1160°F to 1180°F.

Development For Plant-Scale Installation

The development program for the fluid-bed hydrofluorination process included the design of equipment and definition of operating criteria for a plant-scale production system.

After a thorough evaluation, it was decided not to install a fluid-bed hydrofluorination plant at Weldon Spring. Stirred-bed hydrofluorination equipment was already in operation there, and although savings and other advantages would have been realized with the new plant process, it was felt that the benefits would not justify the expenses involved in removing the existing stirred-bed equipment and constructing and installing the new fluid-bed equipment.

Conclusions

Through a comprehensive and extensive program of research, pilot-plant work, and plant-scale activities, Mallinckrodt developed the first continuous-process, integrated fluid-bed system for producing high quality, metal-grade green salt from refined uranyl nitrate as the starting material. The development program was carried on continuously for six years -- from 1957 to 1963.

During the course of the work, Mallinckrodt achieved several major advances in uranium production technology.

Components of the final system, which was capable of successful operation on a plant scale, included: a continuous fluid-bed process for denitration of uranyl nitrate to produce orange oxide; a continuous fluid-bed process for reducing fluid-bed UO_3 to brown oxide; and a continuous fluid-bed process for converting fluid-bed UO_2 to metal-grade green salt.

In comparison with other processes for producing green salt, the major advantages of the integrated fluid-bed process are:

- The mobility of the fluidized solids allows the reaction processes to be carried out in continuous operations.
- Excellent temperature control can be achieved in the fluidized-bed reactions.

- Due to the agitation of the solids, high heat transfer coefficients can be obtained between the solid particles and heating or cooling surfaces.

- Because of the large surface area of the fluidized particles, high reaction rates can be obtained.

All of these factors contribute to making the integrated fluid-bed system more economical, more efficient, and more effective than other green-salt production processes.

Although only the denitration component of the integrated fluid-bed system was used in the plant process at Weldon Spring, it is likely that the entire integrated fluid-bed system, because of its numerous operational and economic advantages, will be employed in any future uranium processing plants that might be built.

Production And Costs

Production and cost details are contained in Section IV.

Related Documents

The following numbers refer to bibliography entries which are the principal sources for more detailed information on advances in continuous green-salt production processes: 8, 10, 16, 30, 41, 46, 54, 66, 67, 80, 103, 166, 169, 170, 173, 199, 201, 222, 223, 224, 228.

Additional references may be found in some of the sources indicated above.

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Section II
Part 11

DEVELOPMENT OF THE DINGOT PROCESS FOR PRODUCING URANIUM METAL

The Uranium Division of Mallinckrodt Chemical Works carried out extensive research and development activities to establish a process for producing uranium metal that would yield purer metal in greater quantities and at lower costs than the reduction and casting process. These results were achieved by Mallinckrodt's development of the dingot (direct-ingot) process.

Background

The first practical uranium-metal production process suitable for use on an industrial scale was the thermite bomb-reduction process developed in 1942 at Iowa State University. (For details, see Section II, Part 1.)

The basic process, which was adapted for commercial production by Mallinckrodt in July, 1943, involved two key steps: thermite reduction, and casting.

Thermite reduction was carried out in a steel bomb shell. First, the shell was lined with a refractory material. At first dolomitic lime, which was relatively expensive, was used as the refractory material; however, development work by Mallinckrodt led to the use of a more practical and far less costly refractory liner -- magnesium fluoride slag, a by-product of the bomb reduction.

After the bomb shell had been lined, the bomb was charged with a mixture of uranium tetrafluoride (UF_4) and a reductant. In the early work at Iowa State, calcium was used as the reductant; however, it was soon replaced by magnesium, which offered several advantages over calcium.

The charged bomb was capped and placed in a furnace where the reduction reaction occurred. During the reaction, the furnace provided the heat required to achieve complete fusion and separation of the reaction products -- uranium metal and magnesium fluoride (MgF_2).

After the reaction had taken place, the bomb was taken out of the furnace and cooled. Then the shell was opened and the products were removed. The solidified uranium metal which was produced was referred to as a "derby."

The second phase of the metal-production process was casting. To produce uranium metal of the size and shape required for subsequent uses, several derbies were melted in an induction heated crucible and vacuum cast in a graphite mold to form an ingot.

In the early work at Mallinckrodt, the reduction furnaces produced approximately 120 pounds of metal per charge. Each derby was about eight inches in diameter by four inches in length. The metal charge for the casting furnace weighed approximately 640 pounds. Each ingot cast was approximately five inches in diameter by 45 inches in length.

Through its development work, Mallinckrodt increased plant capacity considerably. Reduction furnaces were modified to produce approximately 300 pounds of metal per charge. This increased the size of each derby to approximately 12 inches in diameter by approximately four inches in length. The charge for the casting furnace was increased to about 1225 pounds, and this further increased the size of the crude ingots.

Evolution Of The "Dingot" Concept At Mallinckrodt

Early studies conducted by Mallinckrodt to compare the purity of derbies and ingots revealed that the derbies were of purer metal except for a thin layer of impurities concentrated at the surface.

This fact, together with a trend toward lowering non-metallic inclusions in reactor-grade metal, indicated there would be advantages in producing an ingot directly from the bomb-reduction step and entirely eliminating the vacuum casting operation. Hence, Mallinckrodt started an intensive program to develop a direct ingot, "dingot," uranium-metal production process.

Based on the knowledge that only the surface of as-reduced (direct) metal was impure, it was concluded, in planning the initial development work, that larger pieces of the reduced metal should give better percentage yields of pure metal than small pieces. Therefore, larger quantities of green salt were reduced to form larger dingots.

In sum, the dingot concept which Mallinckrodt developed involved: (1) eliminating the melting and casting of uranium metal in graphite crucibles and molds which contaminated the ingot product; (2) producing the final ingot directly from the bomb reduction; and (3) increasing the size of the ingots to produce a product with optimum percentage yields of pure metal.

The Bomb Reaction

Because the dingot concept eliminated the casting step previously used in uranium metal production, Mallinckrodt's development work was concerned primarily with factors involved in the bomb reaction.

The general chemistry of the bomb reaction in the dingot process was essentially the same as that in the derby-production bomb reaction, which is described in Section II, Part 1.

In the dingot reaction green salt blended with chipped magnesium was packed into a steel bomb shell which was lined with powdered magnesium fluoride. The bomb was heated for a specified period in an electric furnace, and then the charge was ignited electrically to start the reaction. Once started, the reaction proceeded spontaneously. Molten uranium metal and magnesium fluoride slag were formed. The uranium, having the higher density, sank to the bottom where it solidified as a large metallic mass. The slag floated and also solidified.

The Development Program

The chief goal of the development program was to establish a dingot process which would produce high-quality, reactor-grade uranium metal in optimum quantities and at minimum costs.

The dingot-metal yield from the reduction operations was dependent primarily upon those operating variables which influenced the length of time that the products of the bomb reaction remained molten. Because of their density differences, the longer the products were allowed to remain molten, the more efficiently did they separate to form a single massive piece of pure uranium metal and a single massive piece of magnesium slag.

Among the variables investigated during the development program were: the size and shape of the bomb shell; heat transfer into and out of the bomb; the relation of the choice of bomb-charge constituents to the evolution of heat during the reaction; the influence of the viscosity of the slag product upon metal separation; the charge quantity; the use of additives; and firing time.

Bomb Geometry

Considerable attention was devoted to the effect of bomb geometry on the quality and quantity of the yield. Bomb geometry influenced heat transfer into and out of the bomb, and heat transfer was one of the fundamental factors affecting the bomb yield.

During the development program, various sizes and shapes of dingots were tried including a 450-pound rectangular one, and cylindrical shapes weighing 900 pounds and 1400 pounds. When the size of the dingot was increased ultimately to 3300 pounds, it was found that the optimum shape for separating the metal from the slag was a right cylinder 18 inches in both diameter and height. This shape was also the best for scalping and extrusion.

The shell consisted of two connected chambers. The upper chamber had a large diameter and contained most of the charge. It was connected by a flared shoulder to the smaller lower chamber, which held the molten metal after the reduction reaction was completed.

The geometry of the 3300-pound dingot bomb offered the slowest rate of cooling possible. In addition, the shape permitted uniform cooling. Another shape might have caused portions of the metal mass to cool faster and entrap slag.

Bomb Preheat

The furnace schedule was another important operating variable investigated during the development program.

Control temperature had a major influence on the yield of the bomb reaction. If the temperature were set too high, the reaction would begin near the charge-liner interface before other parts of the charge had been heated sufficiently to propagate the bomb reaction properly. The result would be poor product separation and low metal yield. A temperature too low also could result in poor yield, or could prevent the reaction from occurring.

The furnace schedule which was finally adopted was a compromise between the high and low temperature extremes. A control temperature of 1150°F was employed for five hours with heat applied in all five heating zones. At the end of five hours, heating of the upper zone was stopped, while heating of the bottom zone continued. This caused the reduction reaction to start in the bottom of the metal cavity. The result was a yield of better than 95 per cent of relatively slag-free metal.

Bomb-Charge Components And Heating Conditions

Another aspect of the development program involved selecting those bomb-charge components which, together with magnesium metal, would provide the most favorable heats of reaction.

Production-grade green salt contained some uranium dioxide (UO_2) and uranyl fluoride (UO_2F_2). Investigation during the dingot development program indicated that very little benefit in heat evolution resulted from the reduction of UO_2 during the bomb reaction. However, investigation did indicate that the UO_2F_2 contained in the bomb charge offered a very definite heat advantage. (In the same sense, it was found that there were some advantages to deliberately adding uranium trioxide (UO_3), which delayed the firing time and also had a high heat of reaction. The use of such additives, however, had to be controlled carefully to prevent too much heat, which could cause a blowout, and to prevent the production of excessive amounts of magnesium oxide (M_gO), which could have a detrimental effect -- as indicated later in this subsection -- on slag viscosity.) Ultimately, it was determined that the UO_2F_2 component -- the water soluble content -- of the charge should not exceed 2.3 per cent.

Slag Viscosity And Metal Separation

Investigation indicated that the presence of too much UO_2, UO_2F_2, uranous uranic oxide (U_3O_8), or UO_3 might prevent good bomb yields.

A variable that was pertinent to this aspect of the development program was the presence of ammonium oxalate insoluble (AOI) in the charge. Because AOI consists of approximately 80 per cent UO_2 and 20 per cent U_3O_8, it has a much lower heat of reaction with magnesium than UF_4 with magnesium.

When Mallinckrodt studied the properties of mixtures of magnesium oxide and magnesium fluoride, the data derived indicated that small additions of MgO to the product slag could assist in lowering the melting point as much as 72°F. Higher concentrations of MgO, however, prevented complete product separation even at temperatures of 3092°F. The presence of undissovled MgO in the slag apparently contributed to increasing viscosity to a point at which it interfered considerably with clean separation of the metallic uranium.

The detrimental effects of the presence of excessive amounts of AOI and MgO led to the establishment of a green salt specification in which the content of pure uranium tetrafluoride was required to be at least 96.5 per cent and the AOI not more than 1.2 per cent.

The Dingot Production Process

Mallinckrodt started regular plant-scale production of crude dingots in 1955. The production procedure, based on the extensive development program, was similar in its basic operations to that employed in derby-metal production, which was the first phase of the older 2-step metal process. The details of the dingot procedure, however, differed from derby production because of the larger unit size involved. The photograph shown in Figure II-11.1 is an excellent illustration of the difference in unit size between a 125-pound derby bomb shell and a 3300-pound dingot bomb shell.

The first step in the dingot metal production process, as in the derby process, was preparation of the bomb shell and liner. A mandrel, as shown in the cut-away drawing in Figure II-11.2, was used to assist in lining the bomb shell. The shape of the mandrel followed, generally, the internal contour of the bomb shell.

After the bomb had been lined, filled and capped, it was transferred to a car that later formed the bottom of the furnace in which firing took place. The photograph in Figure II-11.3 shows a dingot bomb in position ready for firing in one of the car-bottom furnaces.

Firing of the charge was usually indicated by either a sharp rise in temperature as shown on the recording chart of the temperature controller, or by a vibration within the bomb and a characteristic noise. The noise, when transmitted to an amplifier and loud speaker, resembled the sound of violent boiling. It was important to obtain a definite signal that firing had taken place to avoid the danger of a blowout possibly occurring if a bomb fired while it was being handled. A bomb could be removed safely from the furnace for outdoor cooling approximately ½ hour after the charge had fired.

After the bomb had cooled for two to three days, it was moved to the break-out and cleaning areas. The bomb was opened, inverted, and struck with a mechanically operated hammer to remove the lining, product slag, and dingot from the shell. The dingot was dislodged and a mechanical impactor was used to break the large mass of slag into small enough pieces to feed to the slag grinding circuit. The photograph in Figure II-11.4 shows the appearance of slag and a dingot during the break-out operation.

After the large pieces of slag and metal products had been separated, any additional slag adhering to the dingot was chipped away with a pneumatic hammer, and the dingot then was weighed. Figure II-11.5 is a photograph showing a dingot being chipped with the pneumatic hammer.

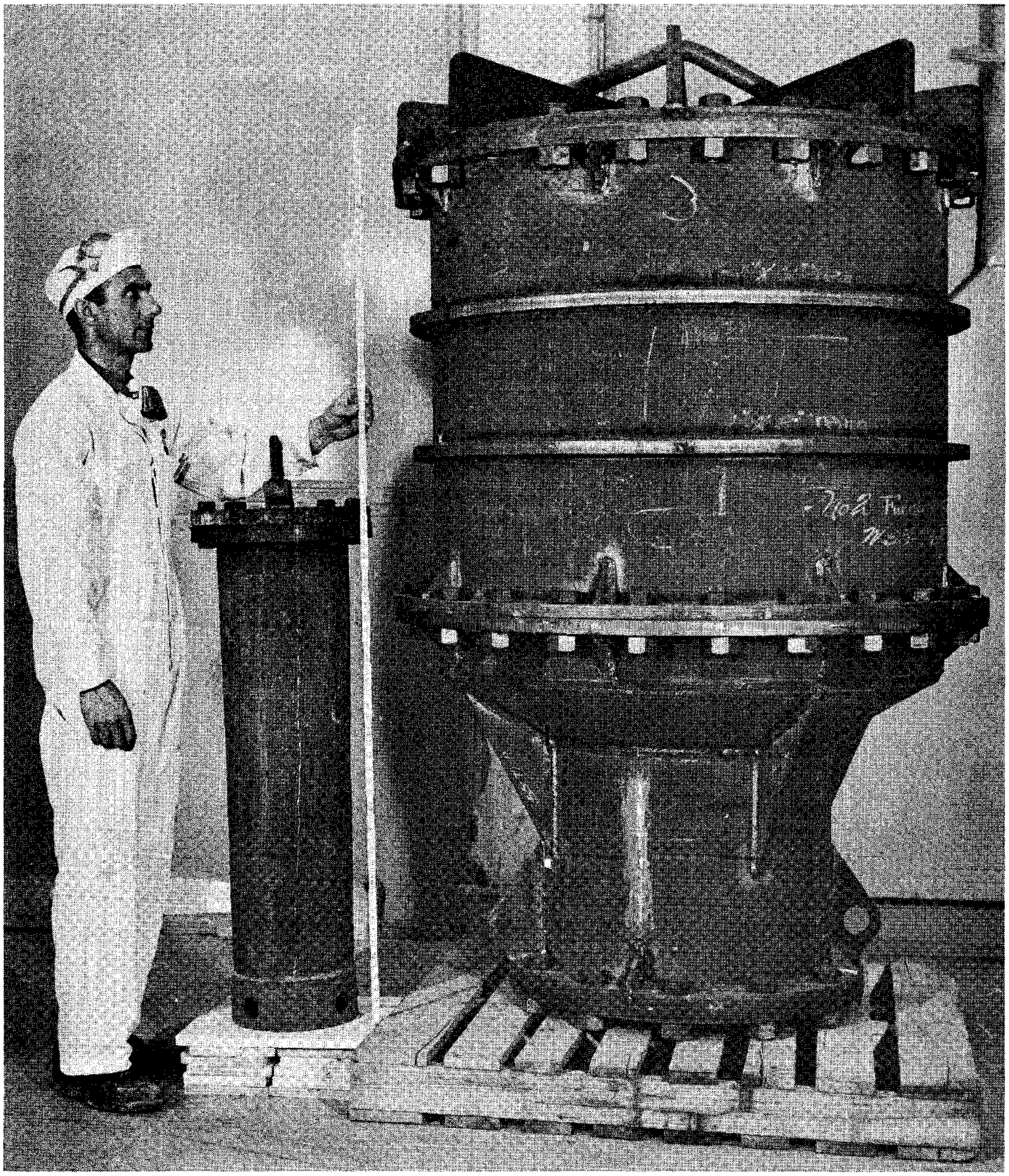

Figure II-11.1 THE CONSIDERABLY LARGER UNIT SIZE OF THE 3300-POUND DINGOT bomb shell is shown here by comparison with a 125-pound derby bomb shell.

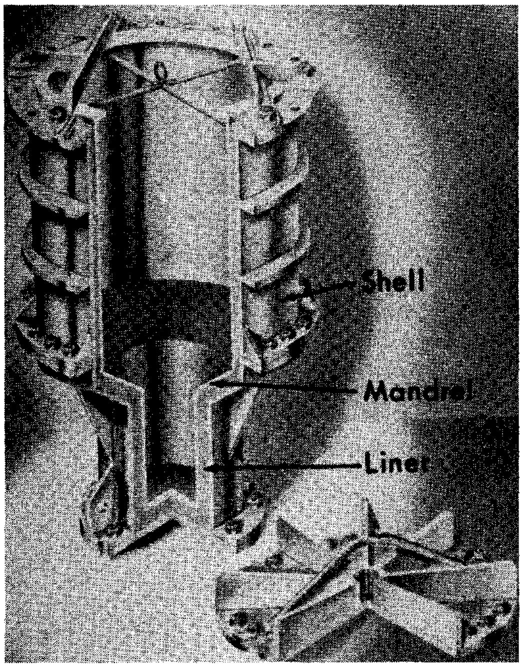

Figure II-11.2 SECTIONAL VIEW OF 3300-POUND DINGOT BOMB SHELL, LINER AND MANDREL.

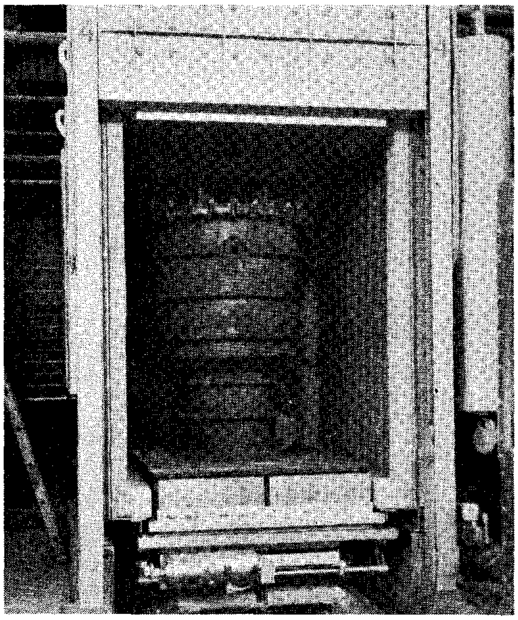

Figure II-11.3 DINGOT BOMB IN CAR-BOTTOM FURNACE ready for firing.

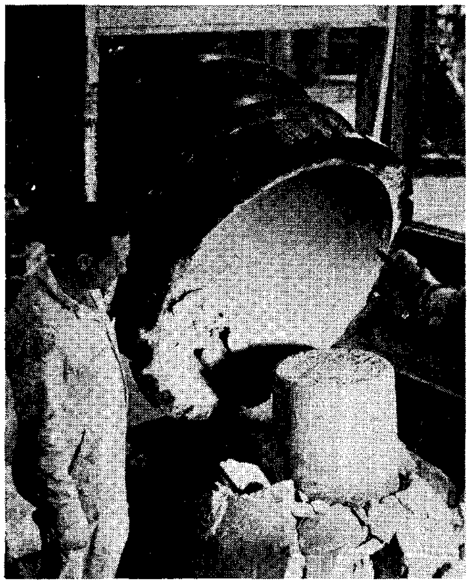

Figure II-11.4 IN THE BREAK-OUT OPERATION, the dingot, product slag and the lining were removed from the shell.

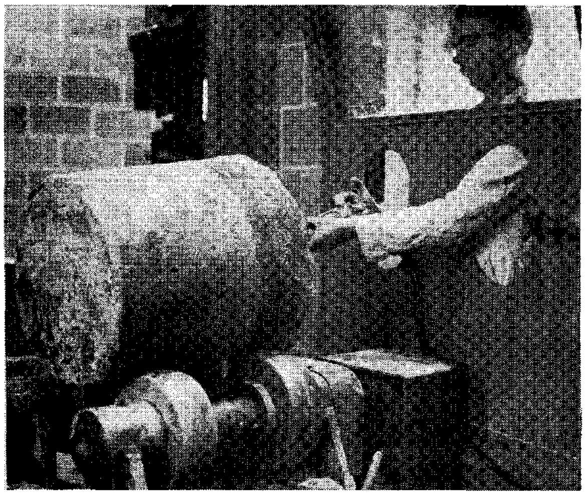

Figure II-11.5 DINGOTS WERE CHIPPED WITH A PNEUMATIC HAMMER to remove any slag adhering to the dingot after the break-out operation.

Figure II-11.6 is a photograph of a 3300-pound crude dingot produced by the process described above.

After the chipping operation, the dingot underwent a "scalping" operation in which all of its surfaces were machined to remove external layers of contaminated metal. The photograph in Figure II-11.7 shows a dingot being "scalped" in a vertical lathe.

Preparation Of Dingots For Fabrication

Dingots produced by the procedures described above measured approximately 17½ inches in diameter by 17 inches in length.

Because of the short length of the scalped reguli, difficulty was experienced in attempting to roll them in the existing rolling mill. The solution to this problem was to add a primary, hot-forming step prior to final fabrication. This also permitted the use of those bomb shapes which produced optimum yields of metal.

With the original concept of the dingot process thus modified, Mallinckrodt started pilot plant studies to investigate the use of press forging for primary fabrication of billets suitable for rolling or extrusion.

Although forging was adequate for the experimental studies, it had certain disadvantages that made it undesirable for full-scale production operations. It was a multi-step, high-cost, low-volume method of production. In addition, forging was not conducive to producing a uniform product. For these reasons, forging was not used for the plant operations. Instead, extrusion was selected.

Extrusion, in contrast to forging, offered the advantages of being a single step, high-volume production process with the capability of producing a wide variety of shapes at moderate cost. Gamma phase (1800°F) extrusion was selected for the Weldon Spring operation because the pressures required were lower than those necessary for alpha phase extrusion.

In preparing the machined dingot for extrusion, it was first sprayed with a phosphate glass-alcohol mixture. Next, it was heated for two hours at 1900°F in an induction coil, after which it was transferred to an inclined table covered with powdered glass. The heated dingot then rolled down the inclined table into the die slide of the extrusion press.

Subsequently, the dingot was extruded through the die to form a uranium-metal rod approximately 13 feet long by seven inches in diameter. The extruded product, which weighed approximately 2650 pounds, was cooled, and subsequently cut to lengths suitable for further processing to slug form. Figure II-11.8 is a photograph of a section of an extruded rod produced by the gamma extrusion process described above.

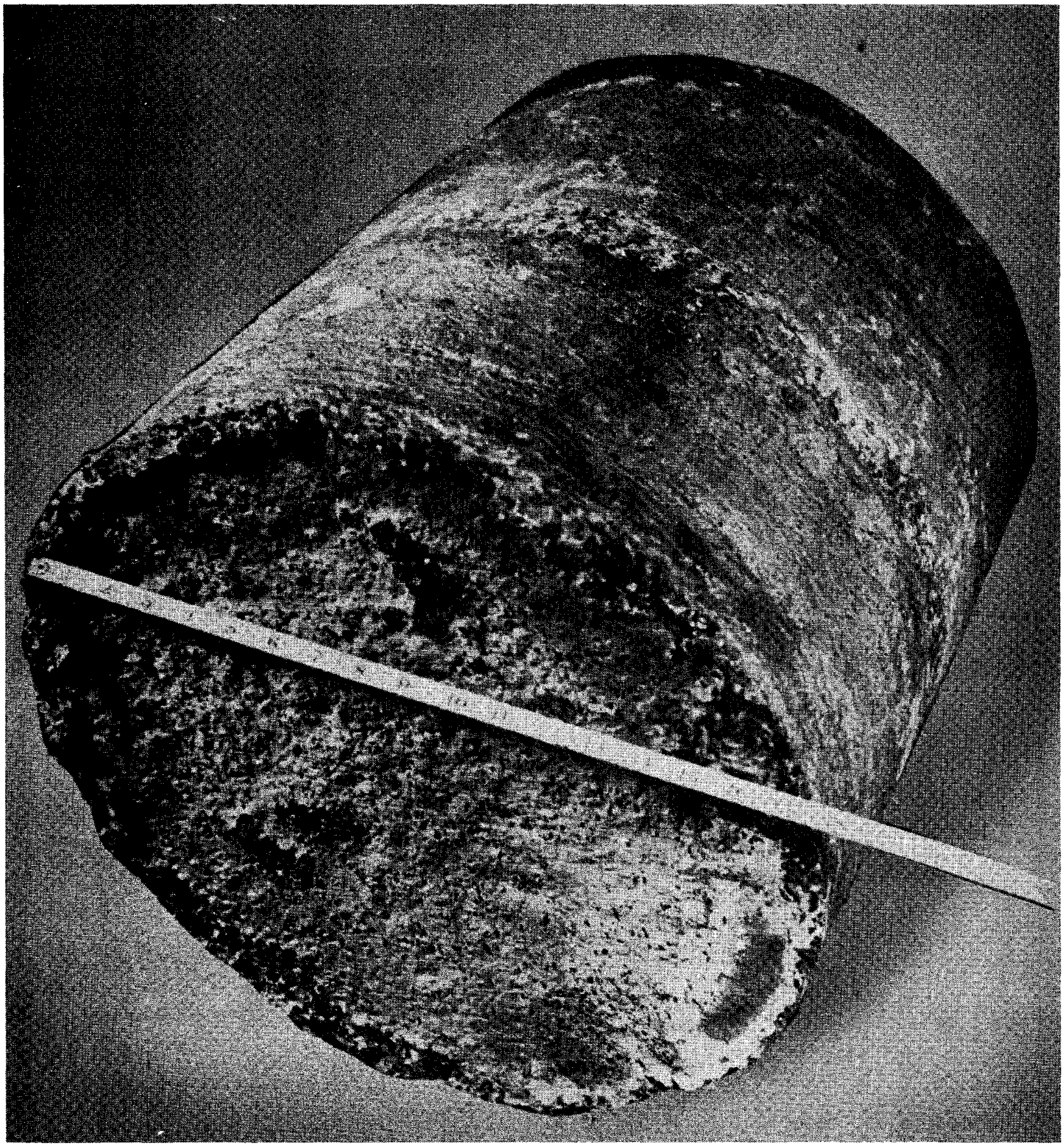

Figure II-11.6 3300-POUND CRUDE DINGOT.

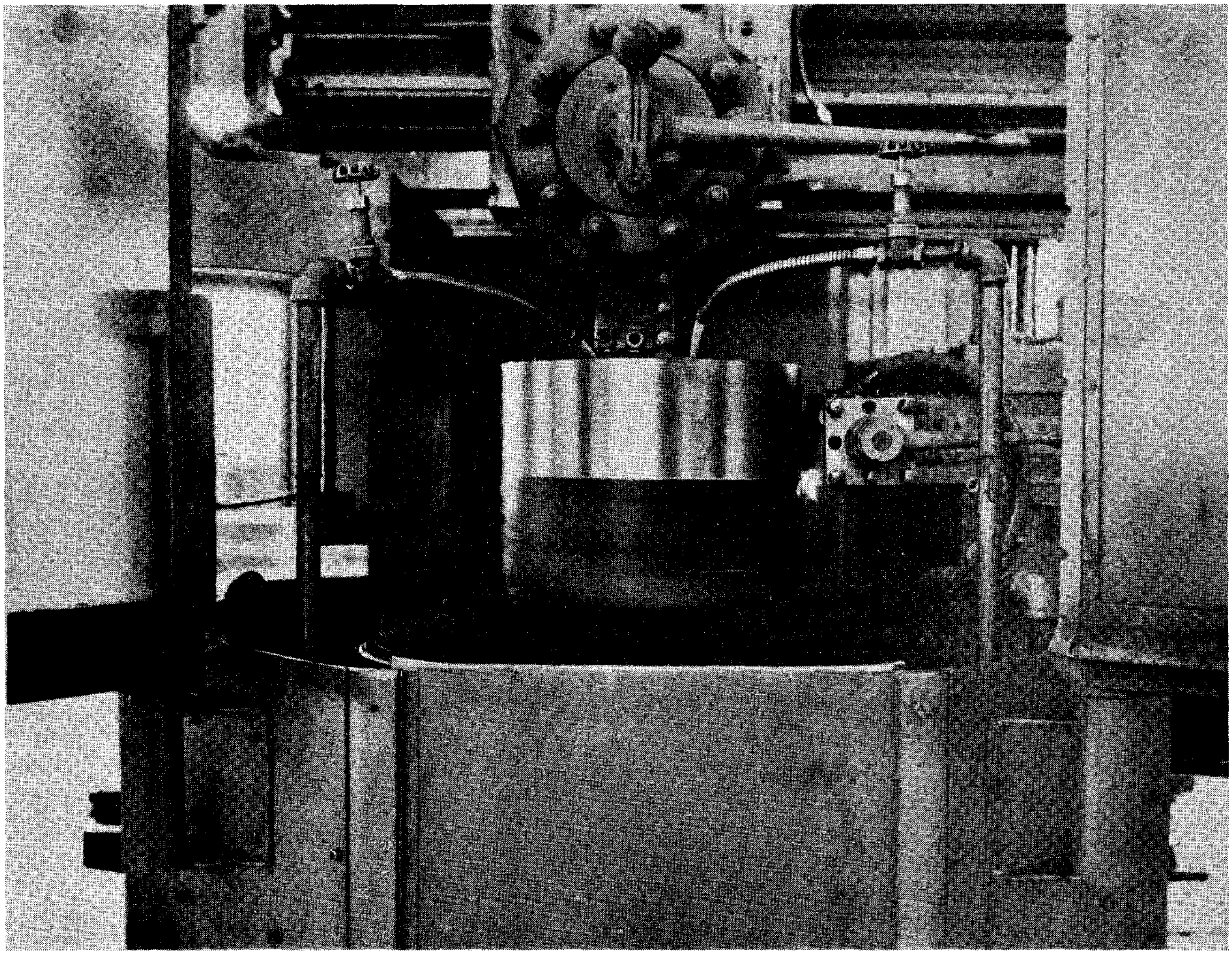

Figure II-11.7 THE SCALPING OPERATION was performed to remove external layers of contaminated metal from the dingot.

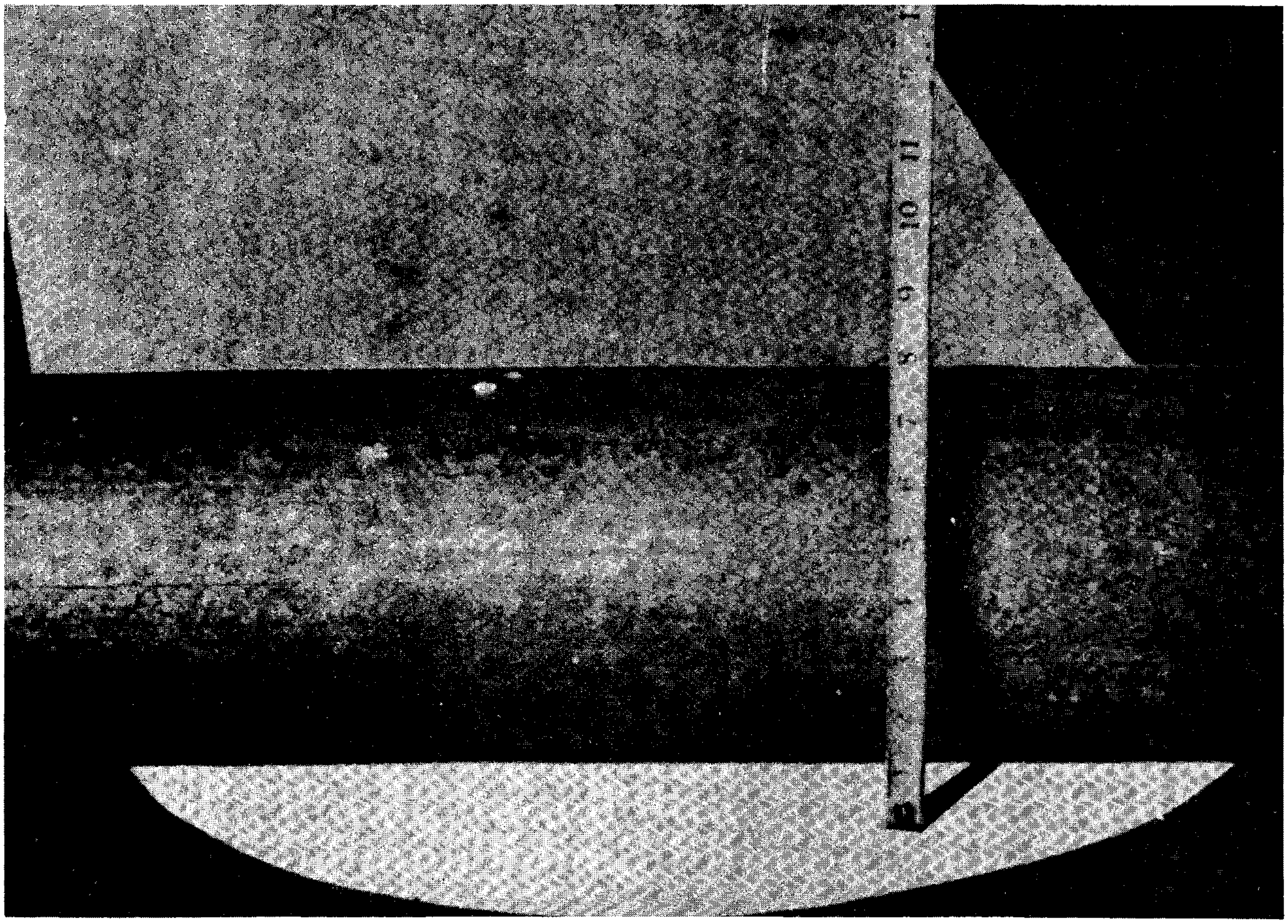

Figure II-11.8 SECTION OF GAMMA EXTRUDED ROD, overall total dimensions of which were 13 feet in length by seven inches in diameter.

Conclusions

Through extensive investigation, research, pilot-plant, and plant-scale work, Mallinckrodt's Uranium Division successfully developed the "dingot" process for producing high-quality uranium metal that is suitable for fabrication to usable shapes directly from thermite reduction.

Production And Costs

Production and cost details are contained in Section IV.

Related Documents

The following numbers refer to bibliography entries which are the principal sources for more detailed information on the dingot-process development work described in this subsection: 10, 13, 16, 21, 29, 32, 40, 41, 57, 63, 76, 86, 89, 97, 119, 120, 128, 165, 173, 187, 199, 201, 202, 208, 222, 223, 224.

Additional references may be found in some of the sources indicated above.

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Section II
Part 12

DEVELOPMENT OF THE ELECTROLYTIC REDUCTION PROCESS

At the time that Mallinckrodt Chemical Works' development work on electrolytic reduction at Weldon Spring was started, the established method of uranium metal production was the multi-step, batch process discussed in Section II, Part 11.

The batch process involves the reduction of uranium trioxide (UO_3, or orange oxide) to uranium dioxide (UO_2, or brown oxide), which is then treated with hydrogen fluoride, an expensive gas, to produce uranium tetrafluoride (UF_4, or green salt). In the next step, the green salt is placed into a refractory-lined "bomb" shell with magnesium (Mg), another costly material. The bomb shell is then heated, and the UF_4 and Mg react to produce a uranium metal dingot. The metal is then rolled, extruded, or forged into usable shapes.

Possible Economies

Electrolytic reduction appeared to offer possibilities of greater economy, because relatively inexpensive carbon and electricity would be used instead of the more costly hydrogen fluoride and magnesium.

Molten-salt electrolysis had been used commercially for many years to produce such metals as aluminum and magnesium from their oxide forms. Further, electrolytic reduction had been employed by Westinghouse to prepare the first uranium metal used in the Manhattan Project.

Background

The early electrochemical work in the atomic energy program included processes at Westinghouse Electric Corporation, Los Alamos Scientific Laboratory and Argonne National Laboratory. All of these processes operated at temperatures below the melting point of uranium. As a result, there were a number of disadvantages. Chief among them were low yields of metal and troublesome reoxidation.

In the low-temperature operations, the deposits of metal on the cathodes were bulky, tree-like structures resembling steel wool. The deposits trapped large quantities of the electrolyte which was difficult to wash free without incurring extensive metal losses. Because of these difficulties, and because the quick success of thermite reduction at Iowa State University offered a practical alternative, work on electrolytic reduction was suspended and not pursued for uranium production until several years later.

In 1957, laboratory-scale work at the AEC's Knolls Atomic Power Laboratory (KAPL) indicated the feasibility of producing uranium metal electrolytically at temperatures above uranium's melting point (2071°F) -- a much higher temperature than used for aluminum or magnesium production.

Development Work At Weldon Spring

With the KAPL results extending the potentialities of electrolysis, development efforts were started at Weldon Spring.

Two major process problems revealed in the KAPL work had to be overcome in the development work at Weldon Spring. These were the settling of the brown oxide into the uranium and the low solubility of the oxide in molten salts.

In addition, a third and even more fundamental problem had to be solved first. The problem was how to contain molten uranium and molten fluorides at temperatures much higher than had been used previously in large-scale electrolytic technology. The solution had to be one which would not result in undue contamination of the product metal.

Careful cost studies, involving reasonable technical assumptions, indicated that if these problems could be solved, a more economical process than the UF_4 - Mg thermite process could be developed.

In the work that followed, Mallinckrodt's initial method employed a graphite hearth for the cathode and a consumable uranium dioxide-carbon anode. The materials for the electrodes were chosen as being the most likely to prevent oxide contamination of the uranium metal product. The approach was successful on a laboratory scale. However, because of the cost of preparing the anodes, attention was turned to developing a direct feed approach. (Concurrently with Mallinckrodt's first work, the Bureau of Mines developed a laboratory-scale process using a rod-type cathode.)

The Production Process

The electrolytic process that Mallinckrodt ultimately developed on a pilot-plant scale employs a cell such as the one shown in Figure II-12.1.

The cell has a graphite hearth, or pot, which acts as the cathode. Direct current is introduced through graphite anodes which are suspended in an electrolyte composed of barium fluoride and lithium fluoride, and containing dissolved green salt and brown oxide. Reduction takes place at a temperature of 2,200° F.

The molten uranium is then vacuum withdrawn from the cell and passed through a heated graphite tube. The pour end of the tube is positioned over a mold within an evacuated enclosure. The casting is either static or, for tubular shapes, centrifugal.

Indicated Advantages

A successful electrolytic process would be likely to offer two chief advantages over the magnesium thermite process. The advantages would be:

(1) Lower cost (probable, but not yet proven).

(2) Uniformity of product, including that produced from recycled scrap.

Conclusions

At the time the Weldon Spring plant was closed, all of the basic problems in developing the electrolytic reduction process had been solved. It is estimated, however, that one to two years of further work would be required to optimize the operating variables and to design a prototype cell in which the economics of the process could be demonstrated.

There is little doubt that if a completely new uranium-processing plant were to be built, the electrolytic process would be preferable to the thermite process for installation in the new facility. Whether it would be advantageous in an existing plant to replace a thermite process with the electrolytic process would depend on many factors, including the outcome of any additional experimental work.

Production and Costs

Production and cost details are contained in Section IV.

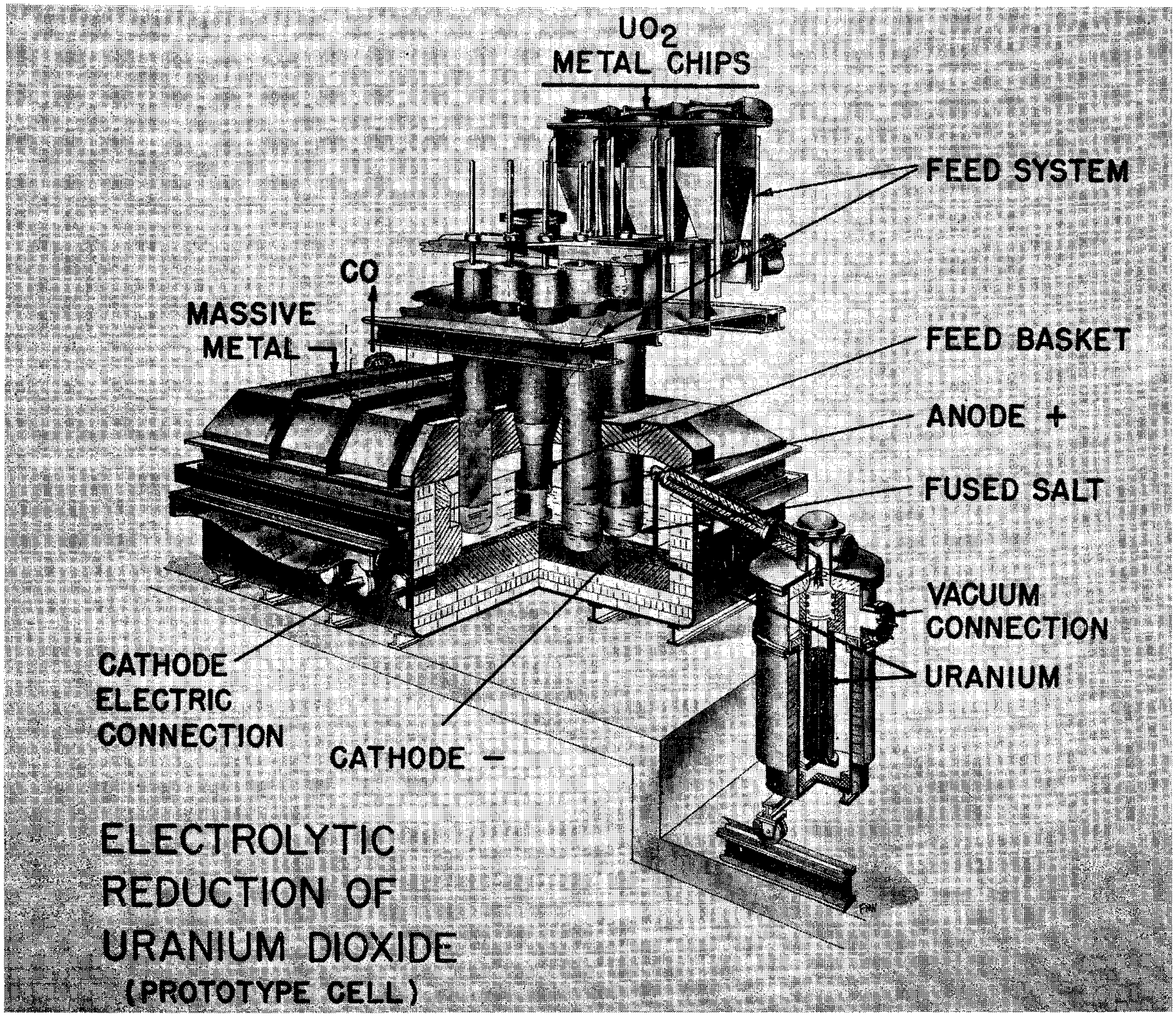

Figure II-12.1 THE ELECTROLYTIC REDUCTION PROCESS developed by Mallinckrodt used a prototype cell, such as this one, for producing uranium metal from brown oxide.

Related Documents

The following numbers refer to bibliography entries which are the principal sources for more detailed information on the electrolytic-process development work described in this subsection: 11, 15, 49, 56, 57, 64, 65, 68, 96, 126, 131, 148, 149, 152, 221, 222, 223, 224, 226.

Additional references may be found in some of the sources indicated above.

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Section II
Part 13

MISCELLANEOUS TECHNICAL DEVELOPMENTS

In addition to the major uranium process developments discussed in detail in the preceding pages, Mallinckrodt Chemical Works was responsible for a number of other important technical advances in connection with the Company's work for the Government.

Sampling, analytical methods development, plant instrumentation, and the adaptation of uranium refining processes to thorium preparation were among the most significant of these additional technical advances. Mallinckrodt's contributions in these specific areas are discussed below.

Sampling

In the field of sampling and sample preparation, the accomplishments included the following:

- In the sampling of domestic concentrates, the Company pioneered enclosed auger sampling which was more reliable than the established open auger method of sampling.

- Mallinckrodt introduced important new concepts and techniques for controlling atmospheric moisture during sample preparation. The control of atmospheric moisture increased the accuracy of prepared samples to a degree not previously achieved anywhere.

- The Company developed the most reliable sample bottling method for uranium concentrates.

- Mallinckrodt invented a practical method for the volume reduction of readily flowable solids having diverse particle sizes.

Analysis

In the field of analytical methods development, the accomplishments included the following:

- The Company developed a rapid and accurate uranium assay method for uranium concentrates.
- Mallinckrodt developed a hot extraction method for analyzing hydrogen in uranium metal. This development enabled a single technician to analyze 100 samples per day as compared to only eight by the vacuum extraction method.
- Mallinckrodt developed a micro-spark method of analysis.
- The Company invented a gas-flow proportional counter for use in electron micro-probe x-ray analysis of elements having light atomic weights.

Plant Instrumentation

The accomplishments in developing instrumentation applications for process control included the following:

- The Company developed a method, and built and successfully operated a unit to analyze the off-gas from the electrolytic cell used for producing uranium metal. The gas was analyzed for carbon monoxide (CO), oxygen (O_2), carbon tetrafluoride (CF_4), and carbon dioxide (CO_2). The problems of high sample temperature varying ambient temperature and corrosion were overcome.
- Mallinckrodt designed and built a portable system to collect process data in digital form on punched tape for computer input. The system recorded the output of a measuring device for any variable which could be expressed as a 3- to 15-pound air pressure on a millivolt signal. In addition to the time of day from a built-in digital clock, twenty other items of information could be recorded in 50 seconds. The console also included calibration equipment.
- The Company designed and built a system to locate and measure the level of metal below an upper layer of molten salt in the electrolytic cell. The measurement was based on the differential conductivity of the metal and salt.

- Mallinckrodt used two systems to safely dissolve massive uranium metal in nitric acid. Such a process can become excessively violent and explosive if the dissolving rate is not controlled. The system that the Company employed at Destrehan Street used feed solution of low acid strength and was programmed on a semi-continuous basis. The system at Weldon Spring used strong nitric acid on a semi-continuous basis. In both cases, monitoring was extensive with provision for automatic shutdown procedures if the process were to go out of control.

- At the Destrehan Street facilities, metal sawdust was reacted with water to form uranium hydroxide. Because the evolution of hydrogen made the process dangerous, extensive instrumentation was provided to control the reaction rate and ventilation. Special provisions were made for an emergency in the event the process were to go out of control.

- Mallinckrodt designed and constructed a system using a very high speed recorder to monitor the speed, position and hydraulic pressure of the extrusion press. Because the time for the entire operation was as fast as five seconds, detection devices were evaluated for response speed and integrated into the system.

- The Company installed a process to collect and neutralize sump liquor automatically for uranium recovery on a semi-continuous basis.

- The Company designed and assembled from standard components an analog computer that permitted rapid determination of the uranium and nitric acid content of extraction feed batches. This device provided a means for determining what volumes of digest, recovery and nitric acid solutions were required to make up a feed batch having a desired concentration and volume.

- Mallinckrodt devised a method for measuring the amount of hydrogen fluoride (HF) in the furnace of off-gas from uranium tetrafluoride (UF_4) production. The gas was condensed and the concentration of HF was determined by conductivity methods using a specially designed cell.

- A "UT-2 Core Tester" that had been developed originally at General Electric, Hanford, Washington, was used to test uranium cores for grain size, cracks, etc. Mallinckrodt carried out development work that resulted in improvements to the machine, and modifications were made for testing many new core shapes.

Development Of The Sol-Gel Thoria Process

In 1963, interest increased in the thorium - 233 U production cycle, and the Atomic Energy Commission directed Mallinckrodt to explore methods for producing a dense thorium oxide suitable for use in such an operation. By the end of 1963, Mallinckrodt had demonstrated a sol-gel process for producing dense thoria on a plant scale.

Subsequent developments by Mallinckrodt included many process improvements and innovations.

The final process employed a fluidized bed denitrator to convert thorium-nitrate solution to a thorium-oxide sol. The sol was dried to gel in pot installations that were used previously for the production of uranium trioxide (UO_3). The gel was densified by high-firing in inductively heated graphite crucibles.

Mallinckrodt produced approximately 500 tons of thorium oxide at Weldon Spring in the first large scale use of the sol-gel process for manufacturing dense thoria.

Related Documents

The following numbers refer to bibliography entries which are the principal sources for more detailed information on the technical developments discussed in this part of the report: 51, 52, 67, 69, 71, 77, 84, 130, 181, 199, 201, 207, 209, 219, 222, 223, 224.

Additional references may be found in some of the sources indicated above.

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Section III

Organization & Management

Section III
Part 1

ORGANIZATION AND MANAGEMENT

The Government's uranium development and production activity operated by Mallinckrodt Chemical Works in the St. Louis area moved through several organizational and managerial phases during the period from its inception in 1942 through termination of the standby contract in 1967.

During the first phase, from April, 1942, until the following fall, the operation was primarily a research project at the Mallinckrodt plant. The chief activities were directed toward developing a satisfactory ether-extraction process for commercially purifying crude uranium-ore concentrates in order to produce highly pure uranium compounds for nuclear reaction experiments.

The second phase was from 1942 to 1945, when the activity -- still at the Mallinckrodt plant -- was operated as a production project. During the earliest stages of production, the only materials produced were uranium trioxide (UO_3, or orange oxide), and uranium dioxide (UO_2, or brown oxide). The "product line" was expanded late in 1942 to include uranium tetrafluoride (UF_4, or green salt), and in July, 1943, Mallinckrodt started its first uranium metal plant.

Major growth in number of personnel employed, scope of work, and physical facilities occurred from 1946 through 1951. The growth period began with the design and construction of the refinery at Destrehan Street, and continued through the subsequent erection of a boiler house, service building, metal plant and green salt facilities.

The decision to build an additional plant at Weldon Spring and to change to a cost-plus-fixed-fee contract required another shift in managerial and organizational emphasis in 1955.

With the opening of Weldon Spring in 1957, the St. Louis-area uranium operations went through a new evolutionary cycle. Operations at Destrehan Street were phased out in 1958. The refinery operation at Fernald, Ohio, was shut down in 1962. Then there was a gradual reduction of the operating scope of the AEC-Mallinckrodt contract until final termination in 1967.

Each of these different phases required changing organizational structures, and managerial plans and programs -- to meet the requirements of the contract.

Background

Mallinckrodt became involved in the first uranium processing because of the Company's outstanding reputation for safely producing high-quality products. These included diethyl ether and high-purity chemicals.

The University of Chicago researchers who were working on the wartime nuclear reactor experiments found that they needed highly pure uranium as fuel for the reactor. They knew that impure uranium concentrates could be purified by extraction with diethyl ether, but that ether was exceptionally hazardous. Dr. Arthur Holly Compton and others from the Chicago team asked several major chemical producers to prepare pure uranium for the project by the ether method, but they all declined.

As Dr. Compton pointed out in his book, Atomic Quest, which relates the story of the American wartime atomic project, the manufacturers who were approached and declined were not only engaged already in war orders, . . . "But also, this was a task they did not want. Ether was not only explosive, but erratic as well."

Dr. Compton explained his reasons for turning to Mallinckrodt:

> "My mind turned to Edward Mallinckrodt, Jr., of St. Louis, whose relatively small chemical works specialized in the production of both ether and pure chemicals. Twenty years before we had known each other well and had spent many hours discussing the factors that might be responsible for the tricky explosions to which ether is subject. Mallinckrodt had a well-earned reputation for caution. Along with this was also a remarkable record for the safety of his employees and a tradition of slow care in making important decisions. We needed his carefulness and his expert knowledge of how to handle ether, but we also needed fast action."

Mallinckrodt's Experience And Qualifications

Mallinckrodt's unique qualifications, including its "tradition of integrity," had been established in its early years, primarily under the direction of Edward Mallinckrodt, Sr., who together with his two brothers, Gustav, and Otto, founded the Company in 1867.

Initially, the Company produced chloroform, spirits of nitrous ether, and an extremely pure grade of the commonly used disinfectant -- carbolic acid. Although the Company almost collapsed in bankruptcy twice during its early years, the brothers would not give in: they ate, lived and slept their business.

No detail escaped their attention. For example, a slightly crooked label on even one small bottle had to be replaced before an order would be shipped. Hard and careful work combined with the Mallinckrodt's excellent business sense sustained and nurtured the Company in its first years.

The brothers' reputation for quality was established early. In May, 1869, a reporter writing about medicines said, ". . . if we must take them, let us have the best . . . from the Messrs. Mallinckrodts' we are sure to get the 'real stuff.'"

In December, 1896, the Company's ninth year, Otto Mallinckrodt died. When Gustav died the following June, the entire responsibility of the business fell to Edward. The Company's progress during the next half century demonstrated his astuteness and ability.

As the firm grew, its business methods, business structure, and product lines kept pace.

In the 1890s, Mallinckrodt began producing a number of new products including: morphine; codeine; hydrogen peroxide; tannic, gallic and pyrogallic acids; sulphon-ethylmethane; and many others which are still important lines today.

During the 1920s, Mallinckrodt established what has become one of its most famous lines -- high purity analytical reagents, which are used in commercial and university laboratories for research and to test the purity of other chemicals. During the same period, the Company developed a major new medium for x-ray visualization of the gall bladder, and also began producing phenobarbital, a product of which it remains the chief supplier.

In February, 1928, Edward Mallinckrodt, Sr., died, ending an illustrious career of 61 years -- all devoted to the Mallinckrodt Chemical Works. In addition to his business achievements, he attained recognition as a philanthropist, having made substantial contributions to educational and other institutions including Harvard University in Cambridge, Mass., and Washington University and St. Luke's Hospital in St. Louis, Mo.

Edward Mallinckrodt, Jr., succeeded his father as Chairman of the Board and successfully carried on the firm's "tradition of integrity." When the younger Mallinckrodt assumed direction of the Company, he already had 24 years of working experience in the enterprise. A dedicated scientist, he had initiated and directed the firm's research which was so successful in greatly improving ether as an anesthetic.

Under his leadership of the Company, quality was given more absorbing attention than before, and in spite of the depression, the decade of the 1930s was, for Mallinckrodt, one of progress and positive change. Especially significant was the addition of a number of Ph.D. chemists who were assigned to improve old products and processes as well as to seek new ones.

The energy, insight and foresight of Edward Mallinckrodt, Jr., was representative of the capability of the entire Mallinckrodt management team. Mr. Mallinckrodt's capacity to grasp new ideas and to throw himself wholeheartedly into new projects was never more obvious than on that important April day in 1942 when Dr. Compton, Dr. Norman Hilberry, and Dr. F. H. Spedding arrived in St. Louis to see him.

The three scientists explained that they urgently needed special uranium compounds in tonnage quantities. The compounds previously had been made only on a test-tube scale, and moreover, the needed material had to have a degree of purity seldom attained even in the laboratory.

Mr. Mallinckrodt's acceptance of the challenge was evidence of his confidence in his company, in his Company's organization and management, in his technical directors, and in other employees upon whose skills success would depend.

His confidence was rewarded. By May, samples of the uranium products which had been requested were shipped for tests, and they met every requirement. Then, overcoming the pressures and handicaps of a wartime situation, the Company designed, procured the equipment for, and built a production-scale extractor. By July -- only three months after the original request -- the Company was producing the needed uranium on a scale of a ton per day.

Dr. Compton recognized the Mallinckrodt organization's important contributions to the atomic energy project -- both technically and in terms of patriotism -- when he wrote, "As long as we have in responsible positions men with such vision, such a spirit of adventure, and such earnest concern for doing their share toward the safety of the nation, we have good basis for confidence in our country's future."

Contractual Arrangements

The initial research project and construction of the batch-type extraction system was conducted entirely upon a letter of intent which resulted in a formal contract only after the delivery of the initial material to the Manhattan District in July, 1942.

The contractual arrangements were made during a visit by Mallinckrodt personnel to the University of Chicago shortly after Dr. Compton and his associates had visited Mr. Mallinckrodt in St. Louis. It became obvious that since Mallinckrodt was to have custody of uranium worth several hundred thousand dollars, and since the Company would be installing a new plant to do the processing, it would be necessary to establish some formal agreement.

Dr. Compton indicated that he had available from the National Defense Research Committee an amount between $10,000 and $20,000 which could be used to support the work. He suggested that Mallinckrodt be paid $15,000 to begin with. On the basis of Dr. Compton's oral assurance that this amount would be forthcoming, the Mallinckrodt team returned to St. Louis to begin work on the project. Shortly thereafter, the Company received a letter of intent pledging the sum of $15,000.

In his book, Atomic Quest, Dr. Compton related, "This was in early May, 1942. Some months later, Colonel K. D. Nichols dropped in at my office. 'A. H.,' he said, 'you'll be interested to know that we have finally signed the contract with Mallinckrodt for processing the first sixty tons of uranium. It was the most unusual situation that I have ever met. The last of the material was shipped from their plant the day before the terms were agreed upon and the contract signed.'"

The initial contract was modified several times during the 25 years of Mallinckrodt's participation in the Government's atomic energy work.

From 1943 to 1946 when the Manhattan Engineering District supervised the activity, the contract was administered on a unit-price basis. Under the agreement, Mallinckrodt was reimbursed for its operating cost and investment in inventory according to a unit price which was based upon the conversion of raw materials to acceptable, specification-grade uranium oxide. When the Atomic Energy Commission took over supervision in 1947, this contractual arrangement was continued and it remained in effect through 1955.

On January 1, 1956, the contract was altered to a cost-plus-fixed-fee type due to the uncertain startup arrangements for Weldon Spring, a growing probability of unusual production demands by the AEC, and the difficulty of establishing raw material quality prior to receipt in St. Louis. The cost-plus-fixed-fee contract remained in effect through June, 1967.

Organization

Mallinckrodt's initial uranium pilot-plant effort in 1942 consisted of approximately 24 people working as a single project group under the immediate supervision of a project manager.

Between 1942 and 1948, the size and scope of the project gradually grew. Research, which had been a significant activity from the beginning, continued to be important; and production which had been somewhat limited at first, steadily increased in importance becoming the project's major function. By 1948, the project employed 250 people and operated approximately $12 million worth of government-owned equipment.

With these changes in its size and scope, the project was re-organized in 1956 into a semi-independent activity which was identified as the Uranium Division. It operated as a separate profit center responsible for certain assigned administrative functions associated only with the uranium work. The parent corporation retained responsibility for the normal business functions not unique to the uranium effort.

The specific functions of the new Uranium Division organization were the following:

- Manufacturing -- The Division operated government-owned equipment to produce purified uranium trioxide, uranium dioxide, uranium tetrafluoride, and uranium metal in the form of reduced reguli or recast ingots.
- Quality Control -- The Divisions' quality control laboratory provided complete specification testing of all raw materials, reagents, and finished product.

- Research -- A small research and development operation for process and product improvement was maintained as an adjunct to the quality control laboratory. In 1949 - 1951, a separate pilot plant and a separate research laboratory were constructed as part of the Destrehan Street facility. The operations there gradually increased in size and capability, and in 1956 - 1957 they were relocated at Weldon Spring.

- Engineering -- The Division established and maintained its own engineering staff to deal with the specific problems of radioactivity and ventilation.

- Dispensary and Health Services -- The Division created and maintained a health department equipped to provide emergency first aid treatment and surveillance of medical examination programs for all Division employees, and to deal with abnormal and potentially hazardous working conditions -- such as the presence of radioactivity and the handling of toxic heavy metals in the form of finely divided oxides. (For additional information on the health and safety operations, see Section III, Part 4.)

- Support Services -- Support services of the Division included: boiler and steam supply -- incorporated into the Destrehan Street facility as a separate installation exclusive of the main Mallinckrodt plant; warehousing; laundry and decontamination facilities, which were unique in comparison to most other Mallinckrodt operations; an extensive library, which contained mainly classified and unclassified technical reports and journals; and an elaborate plant security system to protect and maintain control over data and operations which were classified. (For additional information on plant security, see Section III, Part 3.)

- Administration -- Administrative activities included most aspects of contract administration, cost accounting, and property control.

Because many parts of the operating contract were classified, contract administration was conducted separately by the Uranium Division instead of by the parent corporation.

Under the contract, a fixed unit price was to be paid to Mallinckrodt for each pound of product produced, with the provision for renegotiation based initially upon cost each quarter and subsequently upon cost each four months. To meet the needs of the parent Company's standard cost system, as well as the Division's special need for a negotiation basis, special accounting systems were implemented by an on-site accounting department.

The contract required that Mallinckrodt be responsible for certain quantities of government property. To meet this responsibility, the Division established a property control group which administered effective property control systems.

- Material Accountability -- Because of the high cost of uranium and its extreme strategic value, particular emphasis was placed upon the maintenance of records relating to the location, chemical composition, and assay of all uranium materials under Mallinckrodt's control. The Uranium Division's material accountability group became well known throughout the AEC for the thoroughness of its record systems, for its accuracy and for the several innovations which it initiated. (For further detail on the material accountability operations, see Section III, Part 2.)

Prior to 1956, the organization of the Uranium Division did not include provision for purchasing, general ledger accounting, billing, payroll, personnel, and industrial relations functions. These staff services were provided by the parent corporation.

When the decision was made to construct the Weldon Spring facility, it became necessary to originate a new mode of organization for the Uranium Division. It was determined that to accommodate the cost-plus-fixed-fee type contract, the Uranium Division would have to become a completely separate and essentially autonomous organization, although remaining within the basic Mallinckrodt corporate structure.

On this basis, starting in 1954, the requirements for operating two plants -- Destrehan Street and Weldon Spring -- under one central management group were studied.

An organization plan was developed under which each plant was operated in a line fashion under the supervision of a plant manager with staff groups responsible to a Division Manager who was located at Weldon Spring. New staff functions -- including purchasing, accounting, and administrative services -- which had not been performed previously by the Uranium Division were added.

The two plants were operated and coordinated successfully under this organization plan from 1956, when the water plant and boiler house started operation at Weldon Spring, until the activities of the Destrehan Street facility were terminated in 1958. During the period, the plants effectively met all of the requirements of the AEC. After the Destrehan Street operations were terminated, the organization of the Uranium Division was modified to meet the needs of a one-plant operation.

Beginning in 1956, the Uranium Division prepared organization charts each January and submitted them to the AEC. These charts are on file in the Operations Office at Oak Ridge.

Management Structure

The management history of the Government's St. Louis-area uranium operations can be divided into three distinct periods. During each of the three periods, Mallinckrodt, as a corporation, had official responsibility for management of the uranium activities. Mallinckrodt employed a different management system to administer the work during each of the three periods.

In the first of the three periods, from 1942, until 1950, Mallinckrodt managed the uranium effort as a "project" with a "project manager" directly in charge of all the project activities and personnel. Mallinckrodt selected the project manager, an employee of the Company, with the approval of the selection by the Government. The project manager was assisted by a technical director and sometimes by an assistant project manager.

In day-to-day interaction between the Government and Mallinckrodt, the project manager served as the recognized Company representative for the uranium operation.

Mallinckrodt implemented a second management system during the period from 1950 to 1955. At the beginning of the period, as noted in the above section on organization, the uranium effort was re-organized as a separate Company division -- the Uranium Division.

The chief authority for management of the Division was the "Division manager," a Mallinckrodt employee appointed by the Company with the approval of the Government. In ordinary relations between the Government and Mallinckrodt, the division manager was recognized as the Company representative for the Uranium Division.

Departments were established within the Uranium Division to carry out line and staff functions, as described in the above section on organization, which were unique to the uranium operation. Each of these departments was managed by a department manager who was responsible to the division manager.

Personnel, finance and other ordinary business activities not peculiar to the uranium operation were managed by the parent corporation.

The third management system for the Uranium Division was established in 1956 when the Uranium Division was re-organized to operate as an autonomous unit operating two separate plants.

The new management structure was similar in some respects to the one just described. A division manager was the chief managing authority for the entire uranium activity, and he served as the Company representative in day-to-day relations with the AEC. In addition, department managers supervised and controlled operating departments carrying out specific functions of the Division.

The distinguishing characteristics of the third management system were: (1) Each of the two plants was operated under a separate line-organization management; (2) Staff functions for both plants were supervised by a single, central management group located at Weldon Spring.

At each plant, a plant manager served to direct the line organization functions which included administration and manufacturing. Each plant manager reported to the division manager.

In addition to the plant managers, a single, parallel middle management group was introduced to manage specific staff-organization functions -- administration, personnel, technical development, and plant engineering. Separate managers were in charge of each of these functions, and each manager reported directly to the division manager.

The same basic management structure for the Division was continued after the activities at the Destrehan Street plant were terminated. The only difference was that only one plant line-organization management remained -- for activities at Weldon Spring.

Managers

The initial work on the uranium project, from April, 1942, until Fall, 1942, was carried out under the supervision of Dr. H. V. Farr, technical director. Dr. J. R. Ruhoff was active on the project until 1943 when he was recalled to Army service. Almost immediately, he was assigned by the Army to work on the Manhattan Project.

In the fall of 1942, Joseph Fistere, assistant to the president, became the first project manager. He served in that position until the spring of 1943.

Mr. Fistere, a native of New York City, attended Cornell University until he was called into service in World War I. He joined Mallinckrodt in 1942 after a long and varied career -- including 20 years in China -- with the Allied Chemical and Dye Corporation.

Elected to Mallinckrodt's board of directors in 1945, he was made a vice president in 1947, and in 1949 became president. Even with these increased responsibilities, he maintained a close interest in the affairs of the Uranium Division and took a great personal pride in its progress and activities.

Mr. Fistere retired from Mallinckrodt in 1960.

Harold E. Thayer became the second manager of the uranium operation serving from the spring of 1943 until 1950, and later, from 1955 until 1959.

Born in Rochester, New York, Mr. Thayer, now chairman of the board and president of Mallinckrodt, graduated from the Massachusetts Institute of Technology in 1934 with a BS degree in chemical engineering administration.

Prior to joining Mallinckrodt in 1939, he was with American Cyanamid. His early jobs were in sales and marketing until he was assigned to the Uranium Division, where he began as a construction expediter. He rapidly advanced to plant manager, assistant project manager, and project manager.

During the period when the uranium operation was under his guidance, he had many opportunities to demonstrate his organizational skills.

His insistence on the orderly approach and top performance by "homework," "thoroughness," and "paying attention," soon became by-words. These standards, together with a thoughtful concern for the individual, the helpful criticism for an honest mistake and quick recognition for a job well done, brought about the teamwork which resulted in the expansions, the technological breakthroughs, and the meeting of difficult production goals. Recognition of these contributions came in 1950 when Mr. Thayer was made the youngest vice president in Mallinckrodt's history.

He served as a technical adviser to the United States State Department at the 1958 International Conference on Atomic Energy at Geneva.

Dr. Charles D. Harrington served as manager of the Uranium Division from 1950 to 1955 and from 1959 until the spring of 1960.

Dr. Harrington, an honor graduate of Harvard University -- BS cum laude, MS and PhD in analytical chemistry -- came to Mallinckrodt after six years in the chemical industry. Joining the Company in 1941 he brought top-notch technical capabilities into the organization.

Dr. Harrington was born in White Plains, New York, soon moved to Boston and finally settled in St. Louis in 1941. He became acquainted early with uranium, having studied and prepared uranium hexafluoride in 1940 while at Harvard. This study, incidentally, became a part of the design of the gaseous diffusion process for Oak Ridge.

After a short time in the Company's analytical laboratory he joined the uranium project in 1942. Under his leadership the Uranium Division's technical efforts grew from a handful of people to a well-knit, highly competent technical organization. He continually contributed his tremendous technical abilities especially in times of crises -- of which there were many -- and on numerous occasions personally guided the long, laborious planning and designing of new plants like the green salt and dingot processes.

He is a co-author of the book "Uranium Production Technology," and the 1960 recipient of the American Chemical Society's Midwest Award. He became a vice president of Mallinckrodt in 1960 and in 1961 transferred to United Nuclear Corporation of which he is a vice president and director.

Stanley H. Anonsen was manager of the Uranium Division from 1960 to 1961. For the ten years prior to his appointment as division manager, he had served as assistant division manager.

Mr. Anonsen, a native of Minneapolis, Minnesota, joined Mallinckrodt in 1943.

Starting as a chemist in the laboratory, his ability to organize and get things done soon singled him out as a competent administrator. He figured prominently in handling the growth of the administrative functions of the Uranium Division, first at the Destrehan plant and then in smoothing out the start-up difficulties at Weldon Spring.

Through the years one of Mr. Anonsen's prime responsibilities was to develop and administer cost control procedures. He thus played a key role in the Uranium Division's achieving its enviable record of steadily decreasing costs.

His progress has been most marked. In the Uranium Division, he advanced from chemist to staff assistant, to assistant manager, and then manager. Returning to the parent corporation, he became corporate vice president and general manager of the operations division of which the Uranium Division was a part.

In September, 1961, William J. Shelley was appointed division manager, and he continued in the post until August, 1966.

Mr. Shelley, a native of Wichita, Kansas, was called out of college to serve in the Army during World War II. After three years in the service, he returned to the University of Michigan where he received his BS degree in 1948 and MS degree in 1949 in chemical engineering.

Mr. Shelley came to the Uranium Division directly from school and started his industrial career as an administrative aide. Later his training as a chemical engineer was employed while assigned as a process engineer on plant problems. This experience plus Mr. Shelley's inherently careful engineering approach to administrative problems resulted in steady promotions.

After a period of general administrative responsibilities, in 1955 he was appointed production control manager, responsible for the critical task of getting ores in and finished products out. During this period the Uranium Division developed a materials control organization that became an example of efficiency and thoroughness throughout the Atomic Energy Commission.

He advanced to the position of director of administration in 1960, and assistant division manager in 1961. He was elected vice president of the Company in 1964. In 1966 - 1967, he provided supervisory continuity in many aspects of the activities associated with closing out the Uranium Division.

In August, 1966, Edward Monaco was appointed manager of the Uranium Division, and he continued in the post until the Company's contract with the AEC was terminated in June, 1967.

A native of Bristol, Pennsylvania, Mr. Monaco studied from 1939 - 1943 at the University of Michigan where he earned his BA degree with a major in chemistry. In 1943, he joined Linde Air Products Company, Buffalo, New York, and served there until 1946 as supervisor in the firm's uranium production plants.

He joined Mallinckrodt as a production engineer in September, 1946. From that time until final close-out of the Uranium Division's operations in 1967, his career with the Company was devoted entirely to the uranium processing operations for the Government.

During his nearly 21 years of uranium work at Mallinckrodt, Mr. Monaco served in various administrative and supervisory capacities which involved him in almost every aspect of the Company's uranium-production activities. By 1965, he had advanced to assistant division manager and operations manager.

Upon promotion to division manager in August, 1966, Mr. Monaco became responsible for handling the many difficult management problems associated with the organizational and operational shut-down of the Uranium Division. He was concerned with phasing out production operations, placing the Weldon Spring plant in standby condition, transferring and relocating personnel either to the parent Company or to positions with other firms, and terminating labor contracts. In conjunction with Mr. Shelley, he directed the standby operations at Weldon Spring through a site representative until termination of the contract.

Related Documents

The following numbers refer to bibliography entries which are the principal sources for more detailed information on organization and management as discussed in this part of the report: 3, 6, 9, 11, 13, 14, 15, 16, 19, 23, 24, 25, 36, 39, 41, 50, 58, 59, 61, 75, 81, 82, 98, 121, 164, 173, 190, 191, 192, 195, 197, 198, 199, 201, 203, 210, 214, 215, 216, 217, 222, 223, 224.

Additional references may be found in some of the sources indicated above.

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Section III
Part 2

MATERIAL ACCOUNTABILITY

From the earliest days of its involvement in uranium work in 1942, Mallinckrodt Chemical Works was responsible, under its contract with the Government, for accounting for certain Government-owned materials which had not only important monetary worth but, even more significant, extremely high strategic value. Accounting for the uranium also served as a security measure to detect and prevent diversion to unauthorized sources.

Background

Accountability can be described in its simplest form by comparing it with a bank balance. A certain amount of money is in an account at the beginning of a month, deposits are added and withdrawals subtracted, leaving an end-of-the-month balance which can be checked by physical count.

In money accounting it is not difficult to obtain a perfect balance, but in uranium accountability, a perfect balance is seldom achieved because of uncertainties in measurements.

Accountability In The Early Years -- 1942 - 1946

In the early years between 1942 and 1946 the feed materials were black oxide and sodium uranate, and although grossly impure, they were fairly uniform in particle size and composition. The uncertainties in the measurements of uranium content were small.

End-of-the-month physical inventories for preparing a material balance consisted primarily of a piece-count of feed materials and products, and the sampling of in-process solutions. In the initial plant, the in-process solutions were contained in eight tanks, the largest of which was 500 gallons. The solutions were fairly pure and contained little or no solids, making sampling, sample preparation, and sample analysis a fairly simple task compared to the inventory techniques practiced in the modern plants constructed later.

As production facilities expanded, additional tanks having up to 1000-gallon capacities were installed. New process steps -- the conversion of uranium dioxide to uranium tetrafluoride and the reduction of uranium tetrafluoride to metal -- were also added. These changes added complications to uranium accountability and required additional efforts and knowledge of material measurements.

Plant Expansion Requires Improved Accountability Systems

In 1946, with the start-up of the new Destrehan Street plant built primarily for processing pitchblende mined in the Belgian Congo, many new and complex accountability problems were experienced.

The much larger plant and more complex processes added to the problem of keeping accurate inventories of the in-process material. As was expected, material balance variations increased considerably. Modern statistical techniques were employed to determine the reasonableness of the material balances and where effort should be applied to produce more accurate data.

Also in 1946, the whole program was transferred from the Army to the newly created Atomic Energy Commission. Although the AEC did not alter drastically the basic accountability requirements, many changes were effected to improve the system. Emphasis was placed on statistical techniques to evaluate material measurements and their effect on the material balances. In addition to verifying inventory and material balances the AEC started investigating the control systems employed by private industry in accounting for valuable materials such as silver and gold.

Development Of Improved Sampling Techniques

In 1951, serious material balance discrepancies were experienced. Statistical analysis indicated that the sampling of pitchblende ores performed at a site near the port of entry was in error. The Raw Materials Division of the Atomic Energy Commission reviewed the data and directed efforts to determine the extent of the error.

It was only after an extensive investigation and the joint effort of Mallinckrodt, the Commission, and the contractor performing the sampling, that the cause of the error was discovered and corrected. The Uranium Division's accountability principles proved sound and were an important factor in this investigation. Since then Mallinckrodt played a major role within the Atomic Energy Commission in developing, improving, and evaluating techniques for sampling uranium materials.

New Accountability Methods For Advanced Processing Techniques

Mallinckrodt continued to develop new accountability techniques to keep pace with advances in production technology.

In connection with the development of the continuous ether purification process, the Company developed a special method for measuring uranium in the extraction columns. Similarly, when the continuous process for converting uranium trioxide to uranium tetrafluoride replaced the original batch-type process, Mallinckrodt developed a special technique for determining the quanitities of uranium in the continuous reactors.

In these cases and others of a similar nature, only careful planning in design and start-up provided accurate data necessary for accountability.

Major Sampling Operation At Weldon Spring

When construction of the new uranium processing plant at Weldon Spring started in 1955, plans included a complete facility for the sampling of all feed materials as well as other material. Many domestic and all foreign concentrates were weighed, sampled, and "analyzed for payment" at Weldon Spring. Only because of its excellent accountability record was this important function in the Commission's program entrusted to Mallinckrodt.

For additional information on Mallinckrodt's accomplishments in the area of sampling, see Section II, Part 13.

Mallinckrodt Contributes To Book On Nuclear Materials Management

Recognizing the need to bring together all of the information relative to accountability systems and the value of such a collection to private industry, the Atomic Energy Commission contracted with a private firm to write a book on the control systems used in the nuclear business. After several years of research and effort, Management Of Nuclear Materials, edited by Dr. R. F. Lumb was published in 1960. For the first time and in a single volume, all of the information about the elaborate control systems employed throughout the nuclear materials industry was available.

Matthew N. Kuehn, who was in charge of Mallinckrodt's material accountability program, contributed the chapter on the material management aspects of "feed materials processing." His text remains an important industry reference on sampling, measurement techniques, statistical control, collection and flow of data, material balance reporting, and inventory procedures in the management of uranium production processes.

Conclusions

Mallinckrodt contributed significantly to the progress that has been made in the management of nuclear materials since 1942. Confidence in Mallinckrodt's systems resulted in extending inventory periods from once a month to considerably longer periods. By statistically analyzing measurement data, it was found that 100% measurements were not required to maintain the same level of confidence in the material balances.

In the 25 years that Mallinckrodt was engaged in the nuclear business, the value of the uranium accounted for by the Company amounted to several billion dollars.

Related Documents

The following numbers refer to bibliography entries which are the principal sources for more detailed information on material accountability as discussed in this section of the report: 13, 14, 16, 23, 24, 41, 51, 53, 71, 77, 100, 101, 121, 164, 175, 181, 189, 201, 222, 223, 224.

Additional references may be found in some of the sources indicated above.

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Section III
Part 3

PLANT SECURITY

Because of the tremendous potential of nuclear energy for both military and peaceful applications, the United States' atomic energy programs, including the refining and production of uranium products, have always been at the heart of the national interest and thus have required the highest degree of security.

During its 25 years of uranium work for the Government and the nation, Mallinckrodt Chemical Works maintained an excellent security record.

Background

In the spring of 1939 rumors started to circulate among scientists about the possibility of a nuclear reaction from the fission of uranium -- a reaction which could be capable of releasing an energy force a hundred million times greater than that of burning coal. This same year scientists voluntarily agreed to stop publishing any data which might be of military interest, with special emphasis on uranium work. By September of 1941, when there appeared a definite possibility that atomic energy would become associated with the war effort, formal control was initiated, and security was tightened.

Thus, from its earliest days in April, 1942, the new uranium work going on at Mallinckrodt's main plant near downtown St. Louis was "hush-hush." Similar secrecy existed in all of the many organizations involved in the atomic energy program -- one site had little or no knowledge of what was going on at other sites. Security was tight everywhere.

The work of the Manhattan Project was probably the best kept secret in America's history.

"Tube Alloy Dioxide" And Other Code Names

When Mallinckrodt formally started the uranium research effort on April 24, 1942, the Company, well aware that the uranium work was secret, deliberately chose the name "Uranium Oxide S. L. 42-17" for the project. The name was chosen to disguise the work as being associated with Mallinckrodt's line of S. L. (standard luminescent) chemicals and, therefore, not special or unusual in any respect. However, it was determined that the use of the term "uranium" was contrary to security regulations, and at the request of Government security officials, the name of the project was changed to "Tube Alloy Dioxide."

Code names assigned by Washington were changed periodically to keep the picture as confusing as possible. Documents looked more like a menu than a tally sheet, containing product names such as orange juice, cocoa, derbies, biscuits, rolls, talcum, and vitamins. The AEC letter shown in Figure III-3.1 lists several of these code names as well as others.

Many of the code names eventually became common in routine uranium-industry language, and some of the terms were so appropriate that they still are in use today. "Green salt," a very graphic description of uranium tetrafluoride (UF_4), is a good example of a currently used term, as is "brown oxide" (uranium dioxide, or UO_2), and "orange oxide" (uranium trioxide, or UO_3).

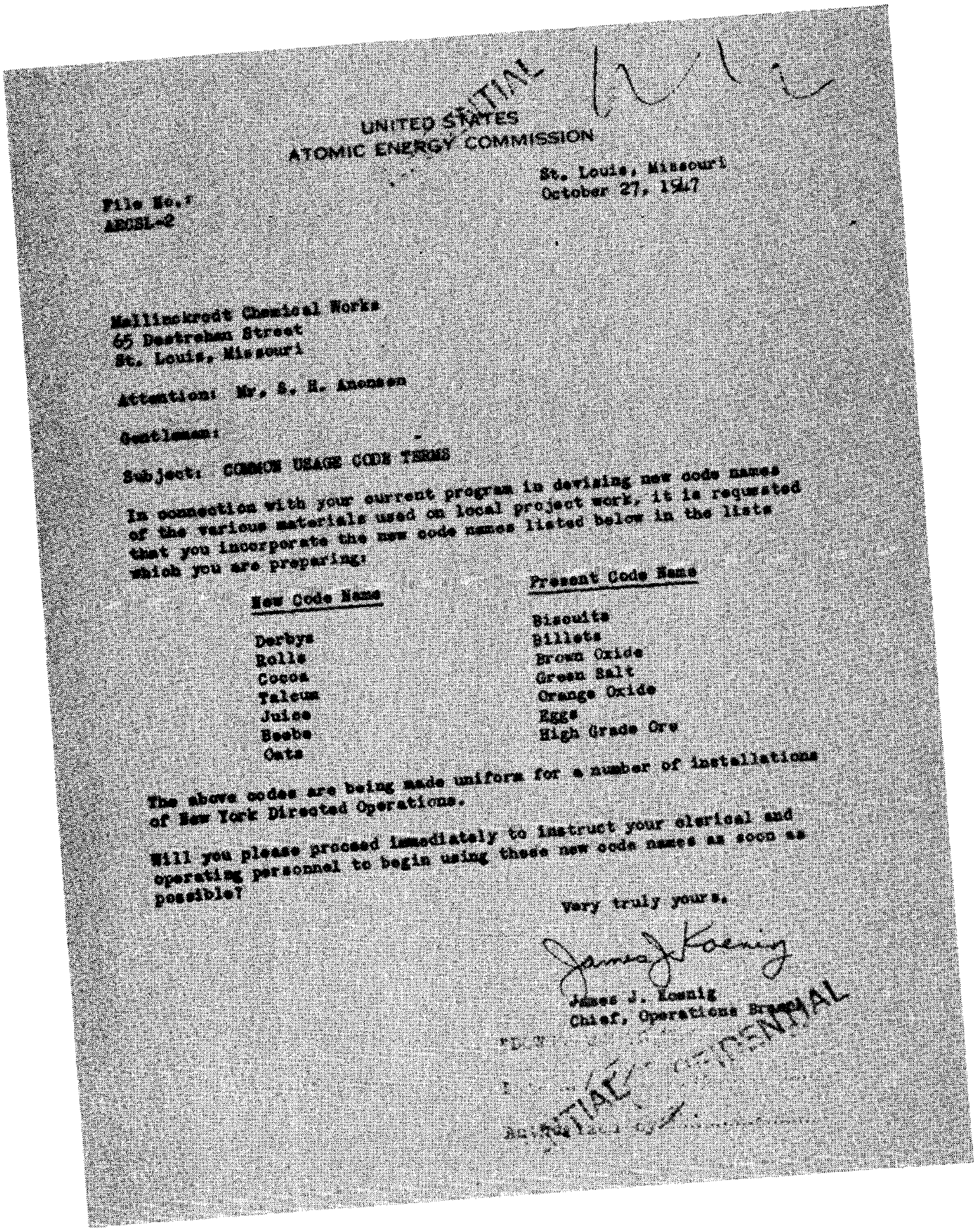
Government Security Control

During the period from 1942 through 1946, personnel background investigations were conducted by Army personnel under the direction of the Sixth Service Command, Chicago. Clearances, in turn, were granted by the Manhattan Engineer District (MED).

During the period from 1947 through 1952, the Federal Bureau of Investigation conducted all background investigations. Clearances were granted by the AEC.

During the period from 1956 through termination of the contract at Weldon Spring, the majority of background investigations involving contractor personnel were conducted by the Civil Service Commission. Clearances continued to be granted by the AEC.

All outgoing shipments were classified and escorted by special couriers who were drawn from a guard complement maintained by the MED in Chicago. Most shipments went by rail, the material and the courier riding in a special caboose which was made a part of regular freight runs. Everything was done to make these runs inconspicuous while, at the same time, providing maximum security.



UNITED STATES
ATOMIC ENERGY COMMISSION

St. Louis, Missouri
October 27, 1947

File No.:
AECSL-2

Mallinckrodt Chemical Works
65 Destrehan Street
St. Louis, Missouri

Attention: Mr. S. H. Anonsen

Gentlemen:

Subject: COMMON USAGE CODE TERMS

In connection with your current program in devising new code names of the various materials used on local project work, it is requested that you incorporate the new code names listed below in the lists which you are preparing:

New Code Name	Present Code Name
Derbys	Biscuits
Rolls	Billets
Cocoa	Brown Oxide
Talcum	Green Salt
Juice	Orange Oxide
Beebe	Eggs
Oats	High Grade Ore

The above codes are being made uniform for a number of installations of New York Directed Operations.

Will you please proceed immediately to instruct your clerical and operating personnel to begin using these new code names as soon as possible?

Very truly yours,

James J. Koenig

James J. Koenig
Chief, Operations Branch

CONFIDENTIAL

Figure III-3.1 CODE NAMES were changed periodically.

Incoming shipments of raw material, pitchblende from the Belgian Congo, traveled by ship from Africa and arrived in New York for processing. The concentrates were then shipped by rail on freight cars. Special cars were used because the material, packed in 55 gallon drums, gave off poisonous radon gas and the cars had to be of the ventilated type.

Offsite warehouse facilities at the Small Arms Plant in St. Louis were utilized for temporary storage of uranium metal and intermediate products. These materials were stored temporarily before being shipped to subsequent destinations: Oak Ridge, Tennessee; Electromet at Niagara Falls, New York, for reduction; or the Simonds Saw and Steel Company in Lockport, New York, for rolling into final shapes for ultimate use at Hanford.

Physical access to the classified working areas was controlled through an exchange badge system. The system was administered so tightly and so successfully that even local inspectors from the fire department and other civil agencies found it impossible to get in.

Security Administration By Mallinckrodt

With the start-up, in 1946, of the new refinery (Plant 6) at Destrehan Street, physical security was provided by Mallinckrodt guards who were stationed on elevated towers from which they could view completely all fence lines surrounding the plant. Each tower facility, such as the one shown in Figure III-3.2, was small -- approximately 4 feet by 4 feet -- and contained a chair, an intercom with guard headquarters, a fan in summer and a small heater in winter.

Mallinckrodt guards, equipped with side arms and riot guns, manned the towers around the clock, seven days a week, for four tense years.

In 1947, the newly formed United States Atomic Energy Commission established offices on the site and government control passed from a military agency, the Manhattan Engineer District, to a civilian agency. At about the same time, a Mallinckrodt security office was established to administer the new Commission's security regulations governing the classification and handling of "restricted data." This was mostly an administrative change, transferring to Mallinckrodt the responsibility to see that educational programs were initiated, manuals were prepared, file cabinets were fitted with combination safe locks, key controls were set up, security checks were initiated, tight rules were made for shipping of classified materials, and visitor controls put into effect.

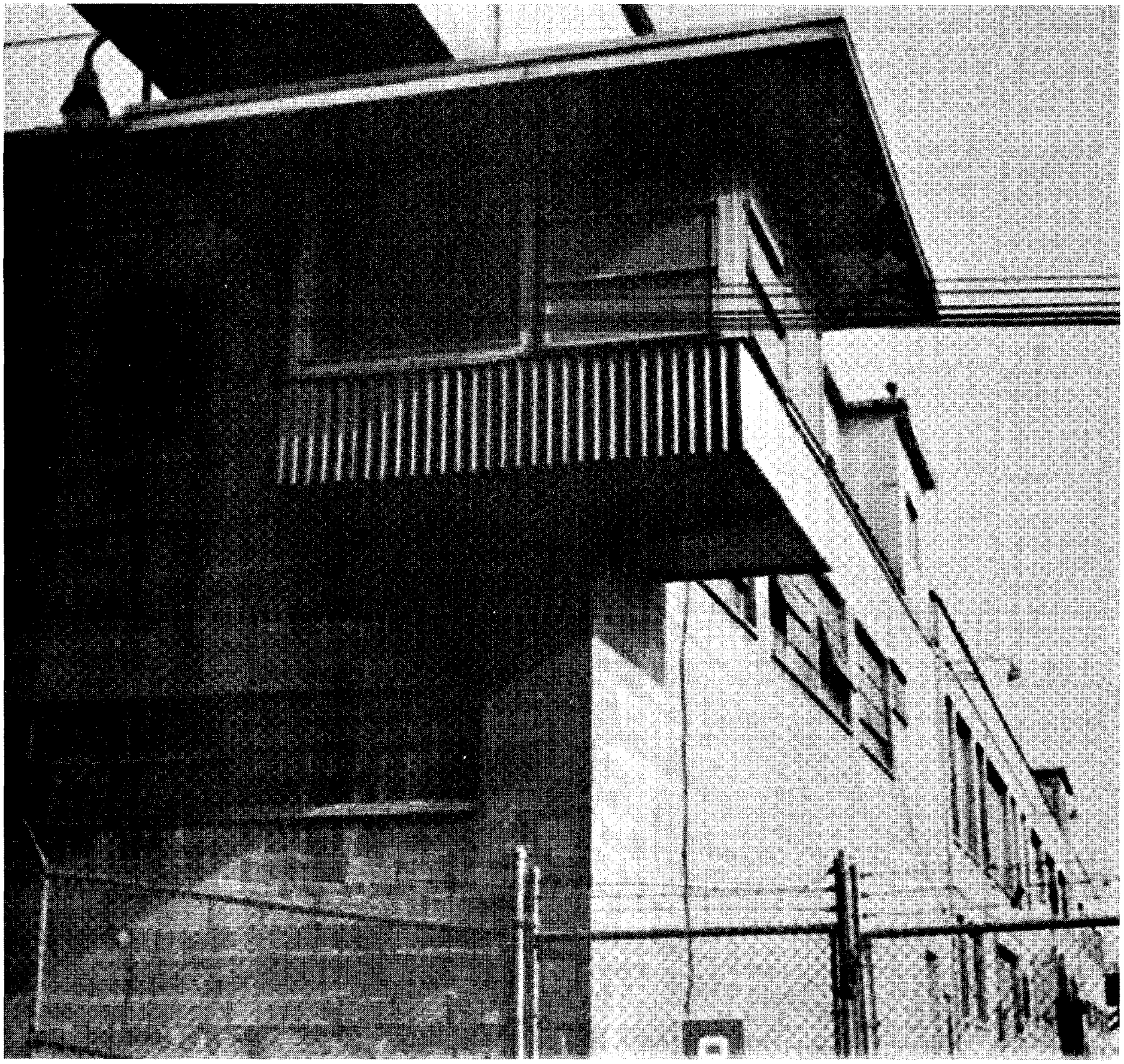


Figure III-3.2 ELEVATED GUARD TOWERS were manned by MCW guards around the clock.

An important part of the new security program, the responsibility for processing personnel clearances of all project employees, consultants, subcontractor people, and some main plant personnel transferred to Mallinckrodt. Everyone working on the project was photographed, fingerprinted and provided with identification badges.

In 1949, plant protection functions became the security officer's responsibility. The project came within Class "A" security category and compliance with stricter government standards was required.

Under Colonel William R. Gerhardt (U. S. Army retired), the uranium activity adopted a further stepped-up security program. He directed the preparation of guard-force manuals, and training and educational refresher courses in use of firearms, operation of the alarm system, fire safety and health physics regulations, property removal procedures, and security control of employees and visitors. One of the most noticeable changes was the increased military appearance of guard force personnel in new uniforms and polished leather.

Additional Security For Increased Plant Operations

In 1949, a building program was completed which provided new offices, a health physics section, a maintenance building and enlarged laboratory facilities. Later, the construction of Plant 6E was completed together with adjoining buildings housing the cafeteria, laundry, employee locker and shower facilities, doctors' examination offices, security office, employee and visitor entrances and guard headquarters.

In 1951, Plant 7 (green salt plant), directly opposite Plant 6, was completed and operations were started up. When Plant 4 stopped processing green salt, the facility was used as a storage area, pilot plant and laboratory.

The security problems involved in protecting Plants 4, 6, 6E and 7 were many and varied, with additional ones caused by the separation of plant sites by public streets. It always meant constant surveillance by guard escort whenever inter-plant movement of classified material was involved. Acquisition of the new site at Weldon Spring in 1956-1957 alleviated many of the security problems experienced in St. Louis.

Plant protection at the Weldon Spring site required a separate guard force organization. As the Weldon Spring operations continued to grow, the other St. Louis operations, including their guard-force functions, were reduced. Ultimately, all of the St. Louis-area uranium operations except those at Weldon Spring were deactivated entirely, and their small, remaining guard-force operations were discontinued.

With the installation of advanced plant-surveillance and communications equipment at Weldon Spring, protection-security operations became almost completely routinized. A communications post, such as the one shown in Figure III-3.3, was the focal point for all security and plant-protection operations.

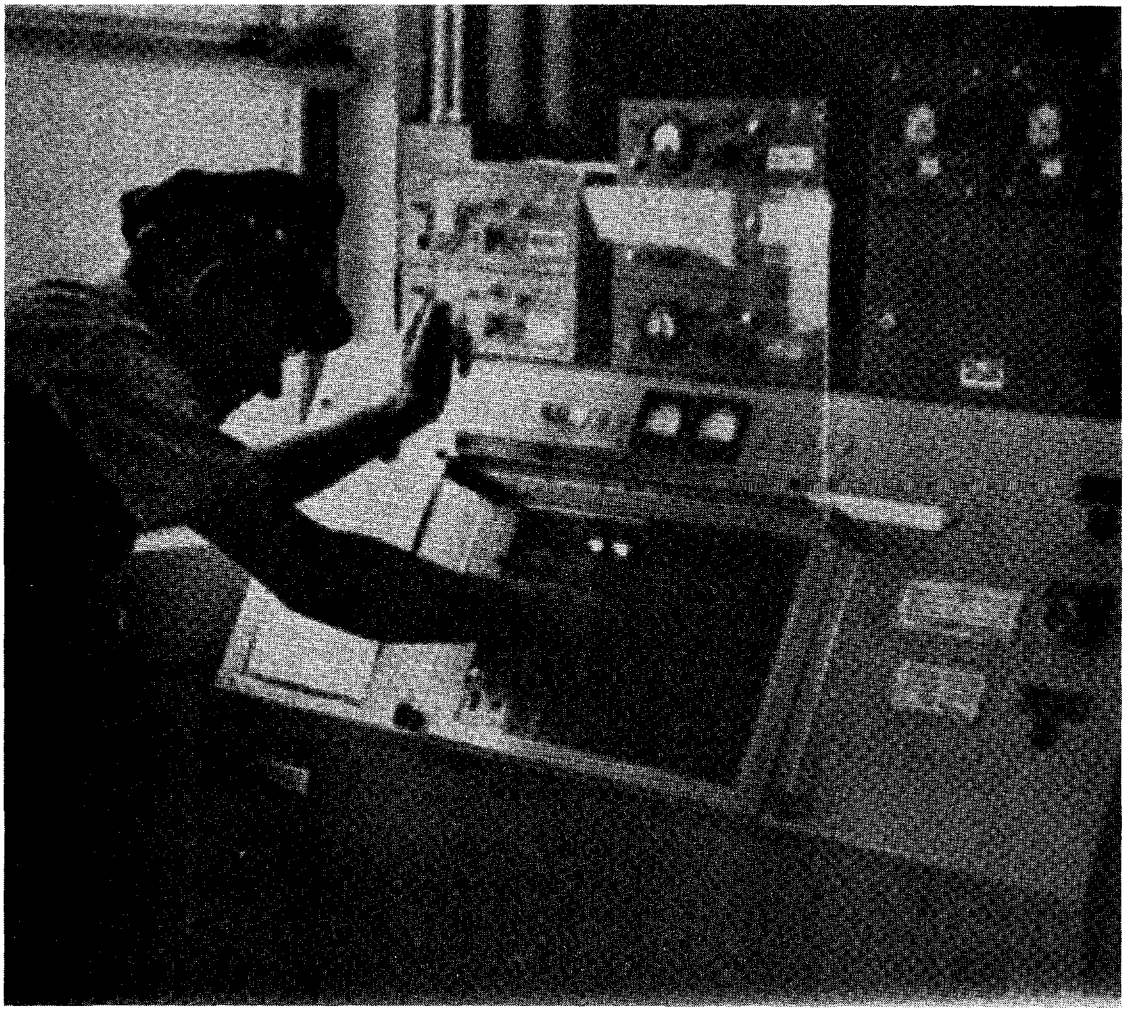


Figure III-3.3 COMMUNICATIONS POST was focal point for plant protection and security.

The panel board shown in the picture contained receivers for ultrasonic alarm systems, as well as fire and evacuation alarms. Other equipment included a two-way radio control unit, emergency telephones and "beeper" devices.

Additional protection was provided through participation in the National Advance Warning System (NAWAS).

Conclusions

From the time that the small uranium research project was started at Mallinckrodt in the spring of 1942 until the Atomic Energy Commission's standby contract with the Company was terminated in 1967, the Mallinckrodt-operated uranium activities in the St. Louis area maintained an excellent security record.

During the entire period -- a quarter of a century -- there was no serious security violation or betrayal of trust which could have jeopardized the national security or proved embarrassing to either the Government or Mallinckrodt.

Related Documents

The following numbers refer to bibliography entries which are the principal sources for more detailed information on plant security as discussed in this section of the report: 14, 23, 24, 26, 42, 73, 222, 223, 224.

Additional references may be found in some of the sources indicated above.

Section III
Part 4

HEALTH AND SAFETY

Throughout the history of the uranium-processing operations in the St. Louis area, vigorous effort was exerted in establishing and maintaining adequate health and safety programs for the protection of people, materials and operations both within plant sites and in off-site areas.

The health and safety work encompassed equipment-design activities, operational discipline, medical programs and audits of possible employee-exposure to uranium toxicity or other contamination, and environmental safeguards.

Background

When Mallinckrodt Chemical Works first undertook the refining and production of pure uranium salts and uranium metal, the entire project was carried out on a short-term, war-time basis under the control of the Manhattan Engineer District (MED).

It was known that uranium was a heavy metal poison such as lead. However, relatively little was known about radiation exposure associated with the material. It was felt that the radioactivity level of uranium was low enough so that small scale, short-term operations would not present a radiation problem.

On this basis, the materials were handled mainly according to standard industry procedures for health and safety protection against the hazards of ordinary toxic chemicals. Precautions of this nature had been basic throughout the history of Mallinckrodt, and the Company's workmen, engineers, and chemists were long familiar with them.

Washington University And University Of Rochester Consulted

Although there was no "formal" health program specifically related to uranium handling when the work was started in April, 1942, there was, nevertheless, a realization from the beginning of a need for specific measures to protect the employees' health from any detrimental characteristics of the uranium.

When Dr. Arthur Holly Compton and his colleagues from the University of Chicago initially approached Edward Mallinckrodt, Jr., to ask for his Company's assistance in preparing purified uranium compounds for use in their experimental nuclear reactor, Mr. Mallinckrodt was concerned about possible health hazards to his employees. He insisted that every measure possible be taken to insure their protection. Therefore, an oral part of the Company's agreement to undertake the project included a provision that the Washington University School of Medicine provide physical examinations for Mallinckrodt employees involved in the work.

In the course of the Uranium Division's history, seven doctors from the Washington University medical-school staff were instrumental in the development of a health program for the Division. One of the seven, Dr. Heinz Haffner, was associated with the health program from 1942 until 1966 when operations at the Weldon Spring facility were terminated. He served as the Division's consultant medical director into the 1950s, and subsequently as a medical consultant. Dr. Norman Knowlton, Jr., also one of the seven Washington University doctors, succeeded Dr. Haffner as consultant medical director.

In addition to the consultation provided by the School of Medicine, other advice and assistance in health and safety protection was provided by the University's Department of Physics and by the MED through its contract with the University of Rochester.

The MED had contracted the University of Rochester to measure the effects of uranium, plutonium, and radium exposure on animals. In addition, the University of Rochester periodically sent men to Mallinckrodt to test dust concentration and radioactivity intensities and to start a film-badge service. In general, the University of Rochester provided specialized health services for the St. Louis uranium operation until the uranium project had its own health department.

AEC Control Places New Emphasis On Health And Safety

As indicated above, during the early years, uranium activities in St. Louis were operated under the MED on a short-term, emergency basis. During the war, Mallinckrodt's contract for the uranium work was renewed at 6-month intervals -- an indication of the "temporary" nature of the project. Because the work was intended to continue for only a short period, radiation exposure was not considered a problem, and no major health programs were required.

At the end of the war, when it was decided that atomic energy was here to stay -- and along with it, uranium processing -- the status of the health and safety requirements changed significantly.

In 1946, responsibility for operation of the uranium project in St. Louis was transferred from the MED to the Atomic Energy Commission. With the passage of the Atomic Energy Act, the Commission assumed obligations for safe operation of its facilities. It was about that time that the St. Louis uranium activity was assigned a permanent status.

This placed an entirely different light on the health and safety aspects of the uranium processing work. Radiation levels that can be tolerated safely for a short period cannot be experienced indefinitely without some risk of damage to health. Also, scheduled production increases would bring increases in radiation problems.

The AEC sent personnel to St. Louis to evaluate the health aspects of the Mallinckrodt operated facility and to make suggestions for possible improvements. The Commission's division of biology and medicine through the health division of the New York Operations Office (NYO), became the outside policy-making group for the uranium project in St. Louis.

Formal Health Program Started In 1947

A formal health program started in 1947 in connection with the addition of the Plant 6 refinery at Destrehan Street and its attendant radiation hazards.

The new health program was started on a crash basis to provide engineering control of health problems, and to establish procedures and regulations for the protection of personnel and materials.

The Plant 6 refinery was constructed to process pitchblende, which contained significant amounts of radium. Because radium gives off gamma rays, which are very penetrating, one of the first projects of the new health effort was to work with engineering to provide thick shield-walls of concrete, brick, lead, or steel around the parts of the process containing radium. Later, uranium ores without radium were supplied, and the thick shielding walls became unnecessary. Another major problem was providing good dust control and ventilation to prevent inhalation of radioactive dusts or gases.

Health Department Established In 1948

In September 1947, following discussions between AEC and Mallinckrodt representatives, Dr. W. E. Kelley of the New York Operations Office submitted to Mallinckrodt a comprehensive outline of recommendations for the initiation of an extensive program to protect personnel from radiation hazards at the project plants.

This eventually led to the health effort acquiring department status in 1948, and during the year, major improvements and large expenditures were made in expanding the health program.

Plant Improvements For Health And Safety

The expanded health program was designed to handle the problems peculiar to large-scale production of uranium metal from all types of feed materials.

The program's immediate objectives were: to install ventilation equipment and shielding, where required, to comply with health protection guides established by the AEC; to educate employees regarding the health-protection aspects of working with uranium materials; and to establish operational procedures for safeguarding the health of employees and for preventing the spread, beyond the plant site, of plant materials possibly exposed to radioactive contamination.

The first efforts of the newly formed Health Department included the tasks of improving conditions in the ore room, developing a dust-control system, and carrying out a series of radiation-shielding, dust-collection and ventilation projects. As the expanded health and safety program progressed, extensive shielding and dust-control facilities were installed in all plants, and those areas which could not be brought under adequate control were replaced with newly constructed facilities.

Figure III-4.1 is a photograph showing a row of giant dust collectors. Dust collection systems were standard items in the plant process in the St. Louis area since 1943-44. The collectors shown in the picture were part of a metal-plant dust-collection system that could move 80,000 cubic feet of air per minute.

Medical And Other Personnel-Protection Services

As part of the expanded health effort, the medical program was stepped up, and other measures were taken to protect personnel and property from excessive exposure to radioactivity and uranium.

All of the facilities of Barnes Hospital and the Washington University medical school were available, and many special tests were conducted there until proper facilities could be set up at the plant. Dr. A. L. Hughes, Professor of Physics at Washington University, served as consultant on the physics of radiation. The expanded health-protection program included frequent medical examinations, and a series of special bio-assay tests, each one designed to measure any biological effects of working with uranium, long before any damage was done to a man's health.

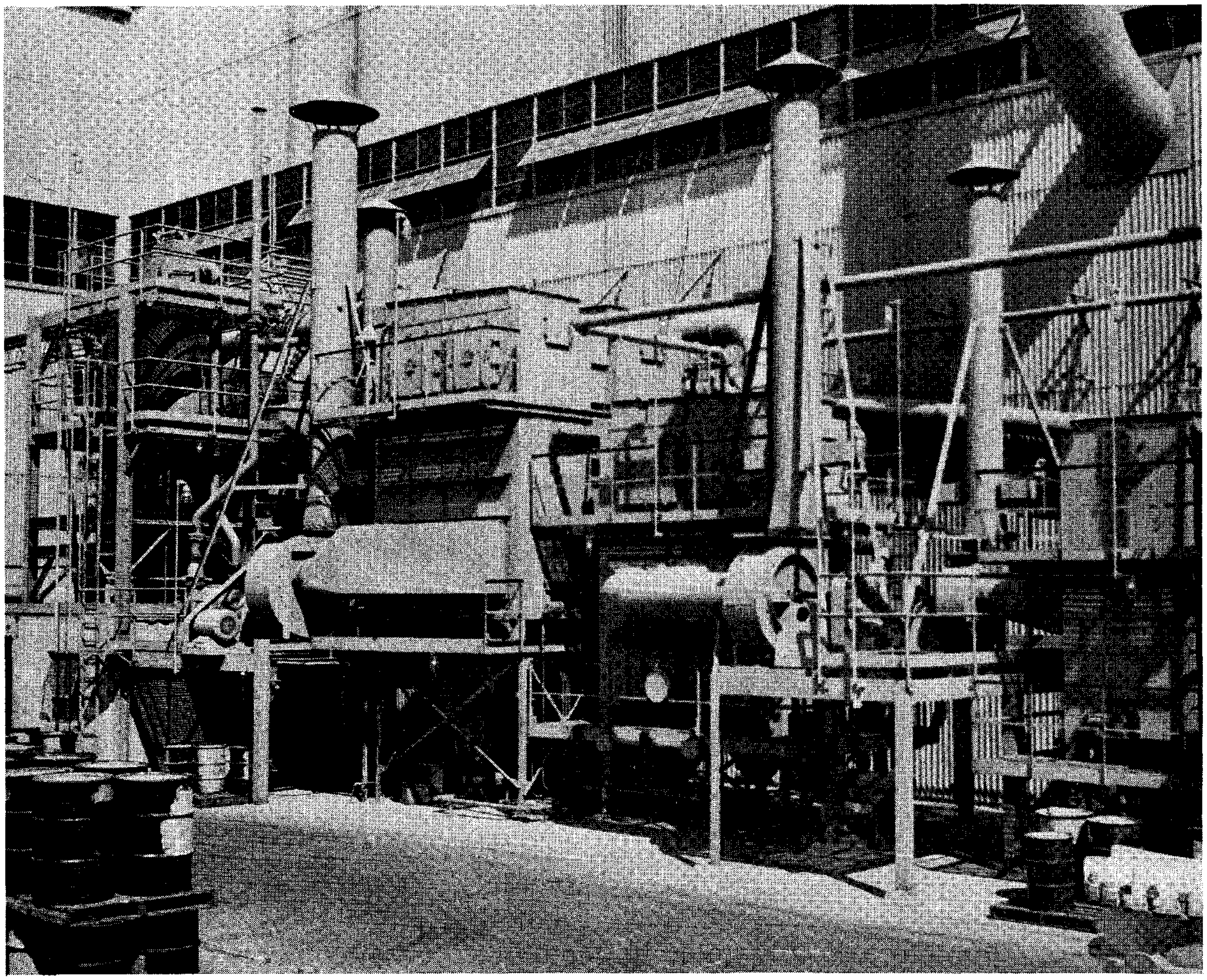


Figure III-4.1 DUST COLLECTIONS SYSTEMS were standard equipment in the St. Louis-area plant processes since the early 1940s.

An additional check on personnel was provided by the film-badge service which had been started in 1942 to measure exposure to beta and gamma radiation. Breath samples, taken periodically from those personnel whose duties involved exposure to radon, were also used to measure exposure to radioactivity and uranium.

Initially, Mallinckrodt forwarded the film badges and breath samples to the AEC Medical Laboratories at Rochester, New York, for examination and checking. Later, the Company had the equipment and technicians necessary for complete and autonomous handling of the film-badge service.

Figure III-4.2 is a photograph showing film badges being checked by a Mallinckrodt employee.

A number of other measures were taken to protect personnel against excessive exposure to radioactivity and uranium. One of the first steps was an employee-rotation program. In addition, a clothing-change and shower program was compulsory for all employees who contacted radioactive materials during their work.

Contamination control was another important part of the health program. It was conducted in accordance with AEC rules to insure that radioactive contamination did not get carried off the site on people's clothing, equipment, trucks, and other objects and become widely distributed about the country.

This program was instigated by the AEC not because there would have been any particular damage done by the materials from the Mallinckrodt operations, but rather as a part of the Commission's nationwide policy, concerning <u>all</u> sources of radioactivity. If radioactive contamination would have been allowed to spread without control, there might have been many serious effects, not specifically on people's health, but rather on various aspects of the highly complex industrial society, such as on the photographic and film industry. Such contamination would also make more difficult and obscure the detection or analysis of fallout or other indications concerning use or testing of atomic weapons.

Special Employee-Relations Activities

Employees who had worked on the uranium project for several years with few formal health regulations found it difficult to understand the new emphasis on physical exams, dust control, radiation shielding, and other health and safety measures.

To explain the program and the reasons behind it, representatives of the health department held many informational and educational meetings with employees for a period of almost two years.

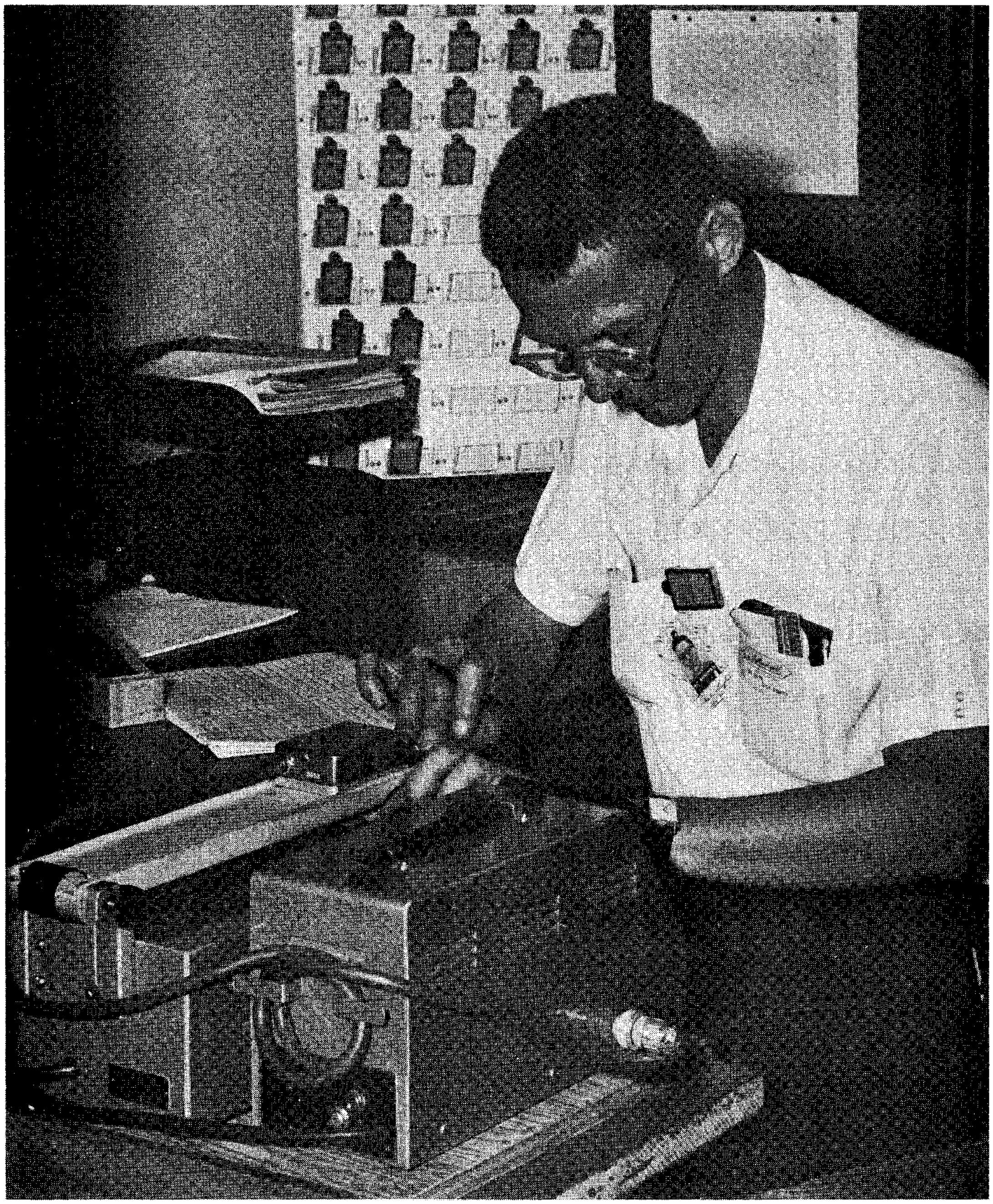


Figure III-4.2 FILM BADGES WERE EXAMINED for signs of exposure to beta and gamma radiation.

Health And Safety Features Incorporated Into Design Of New Metal And Green Salt Facilities

The next major phase of the health program began in 1949 in connection with the design and construction of two new production plants -- Plants 6E and 7 -- to replace the old metal and green salt production facilities at Plant 4, the original plant. The size and other characteristics of the old plant were such that expanded production could not be conducted in a safe and healthy manner while still maintaining a reasonable degree of efficiency.

When the new metal plant was to be built, all of the concentrated experience acquired during the previous two years of wrestling with dust and radiation problems was intelligently applied in developing the design. As a result, when the new metal plant started production, it had only one work station with above tolerance dust concentrations, and this situation was remedied at a cost of less than $100.00.

Soon after the metal plant was completed, design was started on a new green salt plant across the street. The major health advantage in this new plant was the new horizontal-tube screw reactor (see Section II, Part 9) developed by Mallinckrodt, which replaced the old open trays and box furnaces that had been used at Plant 4. Because the new reactors were completely enclosed, it was possible to provide first class dust and radiation protection. This meant that men could go about their normal duties without having to wear respirators and would have to wear them only during dusty jobs when the equipment was opened up for inspection or maintenance.

The plant was also well designed from the safety point of view, especially in view of the fact that hydrogen fluoride, a dangerous and tricky chemical, was involved in the plant operations. Training and excellent attention to safety on the part of the plant operators resulted in this plant winning a safety award for conducting five years of operation without a lost-time accident.

Continuing Improvements To Meet Higher Standards

During the 1950s, the Health Department continued to work closely with design engineers to meet revised standards set by the AEC.

As the nation's atomic energy program expanded, the potential for larger numbers of people to be subjected to long-term radiation exposure led the AEC to introduce a series of reductions in exposure limits. This was done to safeguard not only those directly involved in uranium production and other atomic-energy work, but also, in a broader sense, to protect the nation as a whole as well as future generations.

To meet these changing health and safety standards established by the AEC, as they applied to the Mallinckrodt uranium-processing operations, the Company's engineering personnel had to alter designs of existing facilities as well as construct new facilities.

Control Transferred To Oak Ridge In 1954 -- New Administrative Policy Established By AEC

In 1954, control of the uranium effort in St. Louis was transferred from the AEC's New York Operations Office (NYOO) to the Oak Ridge Operations Office (OROO). With this change, the AEC significantly revised its administrative policy conerning health and safety and other functions of the uranium activity.

Until 1954, the NYOO had participated with Mallinckrodt in all aspects of the health and safety activities, including both establishing standards and implementing programs. In addition, the New York Office served as an important source of technical guidance for Mallinckrodt.

Oak Ridge, in contrast, administered the contract with Mallinckrodt on the basis of an entirely different philosophy. According to the new policy, the OROO acted only in a supervisory capacity. The OROO limited its role to that of establishing health and safety criteria. The contractor was responsible -- on its own -- for implementing activities to meet the established standards.

With the transfer of control to Oak Ridge, the New York Operations Office, no longer in charge of feed materials processing, became the Health and Safety Laboratory (HASL) for the AEC. Until the Weldon Spring facilities were in full operation, HASL assisted Mallinckrodt by performing valuable service functions such as conducting breath-ratings and bio-assays.

Outstanding Health Program And Facilities For Weldon Spring Plant

The continued expansion of the uranium operations in the St. Louis area led to the construction of a new uranium plant at Weldon Spring, Missouri.

The design of the Weldon Spring plant took advantage of both the Company's and the AEC's accumulated wealth of experience concerning the handling of health and contamination problems encountered in uranium processing. The plant layout at Weldon Spring was planned to reduce confusion and lost time in changing clothes and to facilitate control of the spread of contamination. Advanced dust-control systems were designed and installed in the plant, and a modern dispensary and emergency hospital were provided. In general the physical facilities and program for health and safety at Weldon Spring were considered outstanding in the industry.

The Health Department started its operations at Weldon Spring in May, 1957, and because the Department had more office space and laboratory space at the new facility, it headquartered all of its administrative functions, records, and laboratories there.

Existing health programs were continued at both the St. Louis plant and at the new site, and a number of supplemental programs were initiated at the Weldon Spring plant. When the Destrehan Street activities were terminated in 1958, the entire health operation was located at Weldon Spring.

The Health Department experienced substantial growth at the Weldon Spring facility. Two bio-assay laboratories, three medical laboratories for physical examination analyses, and two environmental laboratories were built and staffed.

Safety Program Expanded

Until the change from a unit-price to a cost-plus-fixed-fee contract, safety programs for the uranium activities were administered primarily through the main plant, rather than the Uranium Division. However, a separate Safety Department was created as part of the Uranium Division's industrial relations organization.

In 1959, the Safety Department was combined with the Health Department. The integrated functions were carried out under a single director until final activities were terminated at Weldon Spring.

Environmental Safeguards

In addition to the contamination control efforts mentioned previously, a number of other environmental measurement and control programs were carried out to safeguard the communities -- both local and national -- outside the plant from any hazards which might be associated with the uranium processing. The programs conducted at the Weldon Spring facility to minimize the release of uranium-bearing materials to the surrounding environment were operated satisfactorily during the entire history of the plant.

Plant-process chemical wastes and other process residues were retained in storage facilities located at the site. Treated waste materials were pumped to either the process sewer or to settling basins where solids were removed. The plant-process sewer carried the remaining water effluent into the Missouri River.

Each day, water in the process sewer was sampled automatically to provide continual measurement data concerning any release of uranium-bearing material into the river. Off-site water samples also were collected from other rivers, streams, lakes, and creeks located within the plant's water shed. In addition, air samples collected along and beyond the plant perimeter were tested for uranium and radiation content.

Figure III-4.3 is a photograph showing water samples being collected. Semi-annually an extensive survey of the Missouri River was conducted to determine the content of uranium and other chemical constituents in the river both upstream and downstream from the Weldon Spring plant. Uranium concentrations downstream did not differ significantly from the river's ordinary uranium content upstream from the Weldon Spring facility. Additional checks were made by sampling water at intakes to the water plants of the cities of St. Louis and St. Charles.

In general, sampling of the environment of the Weldon Spring plant indicated that uranium-bearing materials in the area water shed and perimeter air were substantially the same as levels of natural uranium in non-occupational areas, never being more than a small fraction of AEC-accepted limits.

For all shipments of uranium metal and other materials from Weldon Spring, the requirements of the Interstate Commerce Commission were met -- except in cases of shipments that were exempt by some special permit or special provision. Each outgoing shipment or empty carrier was tested and was not released until it qualified under the ICC regulations.

Uranium metal shipped from Weldon Spring was not sufficiently radioactive to require shipping in special containers. Scrap metals from process areas were not sold directly on the open scrap market but were disposed of through licensed handlers.

Conclusions

Health and safety activities continually improved and expanded in scope during the history of the St. Louis-area uranium operation.

The success of the program resulted from the intelligent cooperation and teamwork of all concerned, including Government representatives from the MED and the AEC, Mallinckrodt employees, doctors and scientists from Washington University and the University of Rochester, and many others.

Figure III-4.3 ENVIRONMENTAL MEASUREMENT AND CONTROL PROGRAMS included collecting water samples as part of extensive semi-annual surveys for uranium and other chemical constituents in the Missouri River.

Their combined efforts produced an outstanding health and safety program which was responsible not only for protecting the health and safety of many people, but also for protecting materials and vital industrial and military operations.

Related Documents

A thorough account of the health protection measures employed at Mallinckrodt's Uranium Division is provided in Charles D. Harrington and Archie E. Ruehle's Uranium Production Technology in Chapter 18, "Health Hazard Control."

The following numbers refer to other bibliography entries which are also important sources for more detailed information on the health and safety programs described in this subsection: 1, 2, 10, 13, 14, 23, 24, 27, 33, 34, 35, 37, 38, 47, 60, 72, 85, 123, 124, 125, 127, 174, 182, 183, 188, 193, 194, 196, 198, 200, 201, 202, 203, 204, 205, 206, 213, 214, 220, 222, 223, 224.

Additional references may be found in some of the sources indicated above.

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Section IV

Production and Cost

Section IV

PRODUCTION AND COST

The 25-year history of the production effort under the uranium-processing contracts between Mallinckrodt Chemical Works and the Government can be divided into two major periods -- one, the period from 1942 to 1956, and the other from 1956 until termination of the contract at Weldon Spring in 1967.

During the first period, the work was performed under fixed-price contract arrangements. From 1942 - 1946, operations were located at Mallinckrodt's main plant at Second and Mallinckrodt Streets in downtown St. Louis, Mo. The scope of the project gradually grew, and in 1947, responsibility for the operation was assigned to the United States Atomic Energy Commission. About the same time, the Destrehan Street facilities were added to the project.

During the second period, operations were carried out under a cost-plus-fixed-fee contract. Additional expansion and consolidation led to the erection of the Weldon Spring facility and termination of the Destrehan Street operations.

Throughout Mallinckrodt's work for the Government, production levels and costs related to production levels were classified. This section of the report does not include information that is still classified; however, adequate unclassified information is available to provide the reader with insight into the interrelated facets of operating the production facilities.

Contractual Arrangements

Many aspects of management, organization, administration, operating procedures and Government policies influenced production and costs of the St. Louis-area uranium processing projects. One of these influences was the type of contract under which the work was performed.

During the initial period, under the fixed-price contract, all money exchanges with the Government were based upon a fixed price for accomplishing a specified amount of work. The contract with the AEC provided that 30 days prior to a contracting period, the AEC would inform Mallinckrodt of the quantity and quality of feed materials to be purified during the contract period. At first, the contract period was one calendar quarter. Later, it was changed to one calendar third (four months).

In addition to the basic uranium production operations, Mallinckrodt engaged in research projects and construction projects which the Company carried out for the AEC essentially on a fixed-price basis. Development work on operating processes that were already established was accounted for as part of the manufacturing operation. However, development work on new processes or products was carried as a separate program item, and to secure the contracting officer's advance approval of expenditures, Mallinckrodt was required to define the work carefully in terms of program plans, progress and status during specific phases.

Construction work was subject to somewhat more specific prior definition of details and estimated costs. All construction projects had to be described carefully by Mallinckrodt and approved by the AEC before the work was started. After the construction was underway, status reports and other means were used to keep the AEC fully informed of the progress of the work.

When the cost-plus-fixed-fee contract was implemented and the expansion of the Weldon Spring site started, the policy of before-the-fact approval continued. However, the costs of all programs, including production as well as construction and development work, were tied directly to fiscal-year expenditures. This cost-accounting policy was in accordance with the established administrative procedures of the AEC at the time.

Competitive Atmosphere

Frequently, it is assumed that Government operations are inherently more expensive because they lack the element of effective competition. In the case of the AEC feed materials plants, such a situation did not exist.

During years 1943 - 1947 several plants, in addition to Mallinckrodt, produced UO_3, UF_4 and/or U metal. Production contracts after 1947 were based on competitive bids. Mallinckrodt was low bidder and thus operated the only plant for these three products until the Fernald, Ohio plant was built to meet increased capacity needs.

Beginning with the erection, in 1953, of the Fernald plant, which was operated by National Lead Company of Ohio, step by step competition existed between the St. Louis and Fernald operations. Unlike commercial enterprises, competitive data concerning costs, levels of personnel, and purchasing prices of reagents and materials were made available to the other party. The competitive atmosphere remained intense and unremitting until 1962, and subsequently continued at a somewhat reduced level due to the dissimilarity of operations being carried on at the two plants.

It generally was understood that a similar competitive atmosphere existed between the production reactor operations at Savannah River and Hanford.

Fixed-Price Contract -- 1942 Through 1955

The original contract provided that Mallinckrodt would operate the Government's uranium projects at the Company's facilities in St. Louis. On a regular basis, the Government would supply Mallinckrodt with specified quality feed materials and other items selected by the Government. The Company was expected to provide all of the necessary personnel, reagents and materials required to produce the stipulated amount of specification-quality product. Under these arrangements, therefore, Mallinckrodt invested a certain amount of its own capital funds -- in the form of mechanical and reagent inventories and operating capital -- in the uranium processing work at the Destrehan Street site.

During the period of operation under the fixed-price contract, it was the objective of the AEC to operate the Destrehan Street facility at its maximum capacity with the feed materials then available. Early in this phase of the operation's history, the feed materials consisted primarily of Belgian Congo pitchblende ore. Later, the materials processed included captured German and Japanese concentrates, magnesium precipitates from Belgian Congo and concentrates produced by domestic and Canadian producers.

The amount of income which Mallinckrodt derived from its operation of the Destrehan Street facilities was determined on a regular basis through negotiation with appropriate AEC authorities. A fixed price was negotiated in advance, for a specific period, based on schedule and feed-quality projections. The Company's profit was determined by controlling costs and achieving production schedules.

The estimated cost figures were based on forecasts, for a specific period, of the estimated levels of personnel, reagent consumption, maintenance, and all other similar items required to produce product of specification quality from the feed materials.

The Destrehan Street facility was equipped to handle the Belgian Congo ore. However, processing of this feed material was relatively costly because allowances had to be made for operating a totally enclosed separation plant for protection against the hazardous radium residue present in the pitchblende ore. The concentrated materials, which were secured later, such as the magnesium concentrate from the Belgian Congo, and domestic and Canadian mill concentrates, did not contain radium and therefore were considerably less costly to process than the pitchblende ore.

During the period of operation under the fixed-price contract arrangements, unit prices were established periodically through negotiations with the contracting officer in the St. Louis area. Funding was established primarily on a historical basis; however, major changes in quality of feed materials, in shipping methods and containers, and in prices of reagents, materials and labor were recognized for the coming period, and their influence on costs was taken into account before the fact by responsible AEC personnel.

Unanticipated excessive costs in plant start-up, in yield, or in reagent consumption resulted in reduced Mallinckrodt income for the period. Decreased costs, due to improved procedures and efficiency of personnel resulted in increased income for the period. Normally, the subsequent period was adjusted at the insistence of either party in an attempt to bring the estimates in line with actual costs. The demand for a new product, or the installation of a new process inevitably caused major discussions of pricing methods.

From 1942 until 1948, research and development work was carried out without special funds, and was considered part of Mallinckrodt's routine uranium-processing operations. Initial research was performed in the laboratory and subsequent development work was carried out -- through the final shake-down stages -- in the plant production facility. No pilot-plant facility was available for intermediate development.

Separate, specially funded, major research and development activities began with the establishment of a small laboratory in 1948 to examine the amenability of new feed materials to the dual-cycle ether extraction process which was then in operation at the Destrehan Street plant. This effort was augmented further, in 1948, by the establishment of a green-salt pilot plant for the development of a new green-salt process.

The pilot plant was essentially a reproduction of the various processes in the Destrehan Street refinery. It was used for further amenability testing of feed materials until it became necessary to research the process to be used at the AEC plant at Fernald, Ohio.

Development efforts, which continually grew as the need for uranium increased, included work on the magnesium-fluoride recovery process, the dingot metal process and a forging pilot plant. Machining facilities for forged and extruded materials were established and operated for a period of time.

Cost-Plus-Fixed-Fee Contract

The cost-plus-fixed-fee contract offered many advantages over the fixed-price contract. New production techniques and new products could be introduced without extensive discussion. Construction and development projects could be initiated with more flexibility in design and program than previously.

Production scheduling became a matter of discussion between feed-material contractors and the consumer representatives at Hanford-Savannah River. Production control and accountability procedures provided up-to-date inventory and capacity data so that optimum levels of operation at each site could be established.

Costs incurred at a single site were distributed among all programs at the site -- production, research and construction. Consequently, in any evaluation of operations at a single site, the total level of activity and the distribution of expenditures had considerable influence on the apparent cost of a single specific product or project. Therefore, sites with large amounts or variety of production equipment or research projects were able to show a lower loading of the indirect cost per project than sites, such as Weldon Spring, which had less production capability. Considerable effort was devoted to seeking solutions to these various problems and eventually it generally was accepted that the assignment of a new production or research activity would be based upon the incremental cost of the competing sites.

With commencement of operation of the plant at Fernald and revisions to the Atomic Energy Act of 1954, much of the Government's production activity became classified as "Confidential" instead of "Secret." Subsequently, rather wide dissemination of information about uranium production technology was permitted and Mallinckrodt personnel participated in exchanges of information throughout the plutonium production circuit. Such interchanges promoted better understanding of the influences of cost and quality variations.

Other sections of this report contain detailed descriptions of the technical efforts and their influence on the total AEC production.

Summary And Conclusions

Total expenditures by Mallinckrodt during its 25 years in the atomic energy program are shown in Figure IV. Comparable production data is classified information and, hence, not shown. AEC offices have additional detailed information which is available to authorized persons.

Mallinckrodt operated more than ten years under each of two types of contracts -- fixed price and cost-plus-fixed-fee -- under varying conditions of competition, demand for products, and changing raw materials, processes and products. The Company believes it demonstrated the ability to produce low-cost, high-quality products for the Government under both types of contracts. The Company believes, however, that there is more incentive to improve operations and reduce costs under fixed-price contracts negotiated for short terms. Thus, in Mallinckrodt's opinion, in the long run, fixed-price contracts result in a greater benefit to the Government.

Related Documents

The following numbers refer to bibliography entries which are the principal sources for more detailed information on production and cost as discussed in this section of the report: 3, 6, 14, 15, 16, 19, 23, 24, 25, 36, 39, 58, 81, 164, 173, 190, 195, 199, 201, 214, 216, 217, 222, 223, 224.

Additional references may be found in some of the sources indicated above.

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URANIUM PROCESSING EXPENDITURES
($1,000's)

	Calendar Year	Production	Research and Development	Equipment and Construction	Total
Fixed Price Contract Period	1943	2,721			2,721
	1944	3,147			3,147
	1945	2,329			2,329
	1946	2,619		2	2,621
	1947	5,327		260	5,587
	1948	5,951	45	356	6,352
	1949	5,909	186	581	6,676
	1950	5,984	187	428	6,599
	1951	8,059	154	588	8,801
	1952	8,843	268	511	9,622
	1953	7,943	615	415	8,973
	1954	8,185	854	942	9,981
	1955	9,471	880	1,577	11,928

	Fiscal Year	Production	Research and Development	Equipment and Construction	Total
CPFF Contract Period	1956 (last half)	6,193	446	309	6,948
	1957	12,212	1,214	1,237	14,663
	1958	12,272	1,440	2,805	16,517
	1959	9,891	1,542	2,363	13,796
	1960	10,686	1,304	1,920	13,910
	1961	11,086	1,151	1,924	14,161
	1962	9,869	1,172	1,601	12,642
	1963	8,075	1,134	1,420	10,629
	1964	7,792	1,145	1,116	10,053
	1965	7,156	1,003	717	8,876
	1966	7,363	807	347	8,517
	1967	2,568	86	(291)	2,363

Figure IV TOTAL EXPENDITURES by Mallinckrodt during the Company's 25 years of uranium processing work for the Government. During the period 1943-1956 under the fixed-price type contract, accounting was on a calendar-year basis. During the period 1956-1967, under the cost-plus-fixed-fee type contract, accounting was on a fiscal-year basis.

Section V

Conclusions

Section V

CONCLUSIONS

> "War, peace, freedom --
> each of these we see
> now in new perspective."
>
> --- Arthur Holly Compton*

In April, 1966, the Atomic Energy Commission announced plans to consolidate the uranium feed-materials production operations, at Weldon Spring, and Fernald, Ohio, into one operation at Fernald. The Weldon Spring plant, operated by Mallinckrodt Chemical Works, would be shut down.

Both installations had been operating at reduced levels as a result of production cutbacks announced in January, 1964. Subsequent AEC studies showed that operations could be handled more economically by consolidation.

Production at Weldon Spring ended in December, 1966, and in June, 1967, the final standby status was terminated.

A Quarter Century Of Cooperative Achievement

With the closing of the Weldon Spring plant, Mallinckrodt completed 25 years of continuous service in operating uranium production facilities for the United States Government. During its quarter century of uranium work, Mallinckrodt set records for exceptional achievements in research, process development, and production. In carrying out its contracts, Mallinckrodt played key roles, together with other companies working cooperatively with each other and with the Government, in advancing the atomic age, and in protecting the national security. This joint effort resulted in major contributions to the science and technology of chemistry and chemical engineering, and to a number of other fields including especially technical and engineering administration and management.

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* Arthur Holly Compton, Atomic Quest (New York: Oxford University Press, 1956), p. 289.

In 1962, on the twentieth anniversary of Mallinckrodt's uranium work for the Government, Dr. Glenn T. Seaborg, Chairman of the Atomic Energy Commission, praised the Company for its achievements. In a letter to Harold E. Thayer, chairman of the board and president of the Company, Dr. Seaborg said, "The part that you and the Mallinckrodt people have played in the defense of our country and in the advancement of peaceful uses of the atom has been one of major importance . . . I congratulate all at Mallinckrodt who . . . have contributed so much toward progress in the use of atomic energy."

Technological Progress

During the entire period that the Company engaged in uranium production and development, it strived to achieve three major production goals. These were to improve product quality, to increase production capacity without sacrificing economy or efficiency, and to lower production costs.

Through continued technological progress coupled with effective and creative management, Mallinckrodt achieved all three of these goals. In so doing, it not only fulfilled, but exceeded its contractual requirements.

Fulfilling Other Responsibilities

In addition to its excellent performance in technical and administrative matters, Mallinckrodt's Uranium Division went beyond the provisions of its contracts by accepting numerous civic responsibilities; by engaging in community activities; and by actively participating in national and international professional organizations, meetings and other activities related to the uranium work.

Both the Atomic Energy Commission's and the Company's recognition of the importance of people continued through the final stages of operations at Weldon Spring. When it was announced that the plant was to be closed, every effort was made by the Mallinckrodt organization to absorb employees into other divisions where present or projected needs existed. To make sure that other employees had opportunities to be properly located in good jobs, the Weldon Spring Industrial Relations Department established a personnel placement center which contacted other companies, arranged interviews, and assisted employees in the preparation of resumes.

All of this reflected the concern of both the Atomic Energy Commission and Mallinckrodt for fulfilling social and professional responsibilities which extended beyond the requirements for meeting the immediate material and operational objectives of the plant.

The Future

The achievements that were accomplished at the Government's St. Louis-area uranium processing plants were, without doubt, numerous and great; however, they were just a beginning.

Initially, this nation's atomic energy effort was conducted to meet military and national defense objectives. Later, however, non-military, peaceful objectives for the use of atomic energy were established, and already much has been accomplished to meet these goals. The accomplishments have been achieved through the intelligent, coordinated application of this nation's physical resources and skilled scientific, technical, and managerial talent both in private industry and in the Government. Mallinckrodt, through its uranium process-development and production work, has participated importantly in this effort in the advancement of the atomic age.

In his book, Atomic Quest, Dr. Arthur Holly Compton said, "The high cost of war, now that atomic weapons are in our hands, is forced on our attention with unprecedented sharpness. On the other hand, there lies before us unparalleled opportunity for peaceful growth."*

Atomic energy has practically unlimited potential for advancing mankind.

Presently, fossil fuels -- coal, petroleum, and natural gas -- are the chief sources for the enormous amounts of energy used daily by modern civilization. Because concentrated sources of these fossil fuels "are far from inexhaustible," it is not unlikely that historians of the future will refer to the brief period when fossil fuels were used as "the fossil-fuel incident."

In a speech entitled, "The Atom's Promise," Dr. Seaborg said, "Energy locked in the world's uranium and thorium ores -- which can be used as fuel in nuclear reactors -- is many thousand-fold that in the known reserves of fossil fuels . . ." Dr. Seaborg predicted that "by the year 2000, fifty per cent of all electrical power will come from nuclear sources."

These are only a few indications of the future rewards which are possible -- and probable -- as a result of past efforts in atomic-energy research and development.

Thus, the full implications and significance of Mallinckrodt's uranium-processing work with the Government are yet to be realized.

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* Ibid.

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Bibliography

BIBLIOGRAPHY

Public Documents

1. U. S. Atomic Energy Commission. Standard Safety Requirements. Safety No. 3. April, 1947.

2. U. S. Atomic Energy Commission. Minimum Safety Requirements. Revised edition. Oak Ridge, April, 1958.

3. U. S. Atomic Energy Commission. AEC Contract Policy and Operations. January, 1951.

Books

4. Belle, J. (ed.) Uranium Dioxide: Properties and Nuclear Applications. Washington, D. C.: U. S. Atomic Energy Commission, 1961.

5. Clegg, John W., and Foley, Dennis D. (ed.) Uranium Ore Processing. "Uranium Recovery by Solvent Extraction." Reading, Mass.: Addison-Wesley Publishing Company, Inc., 1958.

6. Compton, Arthur Holly. Atomic Quest: A Personal Narrative. New York: Oxford University Press, 1956.

7. Craven, C. J. Our Atomic World: The Story of Atomic Energy. Oak Ridge: USAEC Division of Technical Information Extension, 1963.

8. Edwards, R. M. The Story of Uranium Processing. A reprint of a talk given in 1956. St. Charles, Mo.: Mallinckrodt Chemical Works, Administrative Services Department.

9. Groves, Leslie R. Now It Can Be Told: The Story of the Manhattan Project. New York: Harper and Brothers, 1962.

10. Harrington, Charles D., and Ruehle, Archie E. (ed.) Uranium Production Technology. Princeton: D. Van Nostrand Company, Inc., 1959.

11. Hewlett, Richard G., and Anderson, Oscar E., Jr. The New World, 1939/1946. Vol. I of a history of the U. S. Atomic Energy Commission. University Park: The Pennsylvania State University Press, 1962.

12. Holden, A. N. Physical Metallurgy of Uranium. Reading, Massachusetts: Addison-Wesley Publishing Company, Inc., 1958.

13. Lumb, Ralph F. (ed.) Management of Nuclear Materials. Princeton: D. Van Nostrand Company, Inc., 1960.

14. Marks, Herbert S., and Trowbridge, George F. Framework for Atomic Industry: A Commentary on the Atomic Energy Act of 1954. Washington, D. C.: BNA Incorporated, 1955.

15. Smyth, Henry DeWolf. Atomic Energy for Military Purposes: The Official Report on the Development of the Atomic Bomb under the Auspices of the United States Government, 1940-1945. Princeton: Princeton University Press, 1948.

16. Thayer, Harold E. The Newest United States Uranium Processing Plant. Paper presented at the Second International Conference on the Peaceful Uses of Atomic Energy. Geneva, Switzerland, June 19, 1958.

17. United Nations. Proceedings of the International Conference on the Peaceful Uses of Atomic Energy. Vol. I: The World's Requirements For Energy: The Role of Nuclear Energy. New York: United Nations, 1956.

18. U. S. Atomic Energy Commission. The Atomic Bonus: Non-Nuclear Benefits from Nuclear Development. Washington, D. C.: U. S. Government Printing Office, 1966.

19. U. S. Atomic Energy Commission. Atomic Energy Facts: A Summary of Atomic Activities of Interest to Industry. Washington, D.C U. S. Government Printing Office, 1957.

20. U. S. Atomic Energy Commission. Progress in Peaceful Uses of Atomic Energy, July - December, 1957. Washington, D. C.: United States Government Printing Office, 1958.

21. Wilhelm, H. A. "The Preparation of Uranium Metal by the Reduction of Uranium Tetrafluoride with Magnesium." Production Technology of the Materials Used for Nuclear Energy. Vol. VIII of Proceedings of the International Conference on the Peaceful Uses of Atomic Energy. New York: United Nations, 1956.

22. Wilhelm, H. A. A History Of Uranium Metal Production In America. Ames, Iowa: Iowa State University of Science and Technology, February, 1960.

Articles and Periodicals

23. _Atomic Industry Reporter: Laws and Regulations_. Section 203: "Atomic Energy Act." Washington: Bureau of National Affairs, Inc., 1955-1959.

24. _Atomic Industry Reporter: Laws and Regulations_. Section 209: "Atomic Energy Commission." Washington: Bureau of National Affairs, 1956-1960.

25. _Atomic Industry Reporter: Laws and Regulations_. Section 221: "AEC Contracts." Washington: Bureau of National Affairs, Inc., 1955-1960.

26. _Atomic Industry Reporter: Laws and Regulations_. Section 246: "Security: Information, Personnel, & Plant." Washington: Bureau of National Affairs, Inc., 1955-1959.

27. _Atomic Industry Reporter: Laws and Regulations_. Section 265: "Health and Safety." Washington: Bureau of National Affairs, Inc., 1955-1959.

28. "The Atom's Promise," (adapted from remarks by Dr. Glenn T. Seaborg, chairman, U. S. Atomic Energy Commission), _Mallinckrodt Magazine_, IX, No. 1 (January, 1964), 16-21.

29. Bradford, William. "Weldon Spring Makes Another Bid For More Metal Business," _Mallinckrodt Magazine_, VIII, No. 5 (December, 1963), 9-11.

30. Brown, Vance. "Fluid Bed System . . . Another Major Technological Breakthrough," _Uranium Division News_, VIII, No. 4 (August, 1963), 17-20.

31. Brown, Vance B. "Impervious Carbon and Graphite in HF Processing," _Chemical Engineering Progress_, LVII, No. 4 (April, 1961), 69-73.

32. Buntz, William. "Gap Closed in Core Plant Facilities," _Uranium Division News_, VI, No. 6 (December, 1961), 14-18.

33. Burr, Richard. "Criticality Control Introduces New Concept of Nuclear Safety," _Uranium Division News_, VII, No. 2 (April, 1963), 8-11.

34. Burr, Richard. "Yes, We Are a Good Neighbor," _Uranium Division News_, VI, No. 3 (June, 1961), 2-4.

35. Caplan, Knowlton. "Top-Flight Health Program Pays Off in Healthier People," *Uranium Division News*, VII, Nos. 3 & 4 (June, 1962), 36-39.

36. "The Chemical World This Week: Research & Technology Concentrates," *Chemical and Engineering News*, (May 7, 1962), 41.

37. Daly, George. "Safety Committee -- the Shortest Distance Between Two Points," *Uranium Division News*, VI, No. 4 (August, 1961), 4-6.

38. Esteep, Ray. "Safety is Like Breathing . . . ," *Uranium Division News*, VII, No. 2 (April, 1962), 22-24.

39. Fain, Pat. "Residues from Uranium Operations," *Uranium Division News*, V, No. 2 (April, 1960), 6-8.

40. Fellows, John. "Hot Off The Press -- Hollow Cores," *Uranium Division News*, VI, No. 4 (December, 1960), 20-22.

41. Ferguson, Gary W. "Weldon Spring, A Key Producer in America's Atomic Program," *St. Louis Post-Dispatch*, April 12, 1959.

42. "Guards Always in Training," *Uranium Division News*, VII, No. 5 (October, 1962), 26-27.

43. Harrington, C. D., and Ruehle, A. E. "Preparation and Properties of Uranium Dioxide Powder," *Chemical Engineering Progress*, LIV, No. 3 (March, 1958), 65-70.

44. Harris, John. "The Magic Key: $E = mc^2$," *Uranium Division News*, VII, Nos. 3 & 4 (June, 1962), 25-31.

45. Hartmann, Robert. "Forge Press Again In Service," *Uranium Division News*, VI, No. 2 (April, 1961), 10-12.

46. Hedley, W. H., and Roehrs, R. J., and Trask, W. T., Jr., "Production of Uranium Dioxide by Flame Denitration," *I&EC Process Design and Development*, III, No. 1 (January, 1964), 11-14.

47. Hoffert, Jeanelle. "1962 Meeting of Health Protection Personnel Held in St. Louis," *Uranium Division News*, VIII, No. 1 (February, 1963), 24-25.

48. Hoffert, Jeanelle. "'These Things I remember . . .'," *Uranium Division News*, VII, Nos. 3 & 4 (June, 1962), 32-35.

49. Hubbard, Keith, and Robinson, Noble. "New Process for Making Uranium," Mallinckrodt Magazine, IX, No. 8 (August, 1964), 12-18.

50. Kelley, Wally. "Facts and Figures -- Unlimited," Uranium Division News, VI, No. 6 (December, 1961), 27-29.

51. Knebel, Al. "New Sample Splitter on Stream," Uranium Division News, VII, No. 5 (October, 1962), 23-25.

52. Kneip, Ted. "Analytical Unit Reorganizes to Give Better Service," Uranium Division News, VIII, No. 4 (August, 1963), 4-8.

53. Kuehn, Matt. "Uranium Division Plays Major Role in Development of Accountability Techniques," Uranium Division News, VII, Nos. 3 & 4 (June, 1962), 44-46.

54. Kuhlman, Carl W., Jr., and Swinehart, Bruce A. "Production of Uranium Tetrafluoride: Thermal Damage Effect," Industrial and Engineering Chemistry, L, No. 12 (December, 1958), 1774-1776.

55. Lehmann, Alan. "Are We a Good Neighbor?," Uranium Division News, VII, Nos. 3 & 4 (June, 1962), 57-61.

56. Leifield, Bob. "Electrolytic Preparation of UO_2," Uranium Division News, V, No. 5 (October, 1960), 9-10.

57. Madsen, Dell. "What's a Patent?" (part II), Mallinckrodt Magazine, IX, No. 11 (November, 1964), 21-25.

58. "M.C.W. Directors Meet at Weldon Spring," Uranium Division News, VIII, No. 3 (June, 1963), 14-15.

59. Miller, Earl, "Our Goals," Uranium Division News, VI, No. 5 (October, 1961), 14-15.

60. Moore, Neal. "World Standards in the Making," Uranium Division News, VI, No. 5 (October, 1960), 24-25.

61. Morgan, Perry. "Why We Are Here," Uranium Division News, VII, Nos. 3 & 4 (June, 1962), 40-43.

62. Petty, William. "Third Information Exchange Meeting Held in Europe," Uranium Division News, VI, No. 6 (December, 1961), 8-10.

63. Philoon, Wally. "Uranium Recovery from Slag," Uranium Division News, VIII, No. 1 (February, 1963), 14-17.

64. Piper, R. D., and Leifield, R. F. "Electrolytic Production of Uranium Metal From Uranium Oxides," I & EC Process Design and Development, I, No. 3 (July, 1962), 208-212.

65. Piper, Roger. "Electrolytic Uranium Grows Up," Uranium Division News, VII, No. 6 (December, 1962), 6-7.

66. Robinson, Noble. "UNH to UF_4 Via the Fluid Bed," Uranium Division News, V, No. 4 (August, 1960), 8-11.

67. Ruehle, Arch. "Development and Technology . . . the Story of Progress," Uranium Division News, VII, Nos. 3 & 4 (June, 1962), 16-21.

68. Ruehle, Arch. ". . . Scrap, Anybody?," Uranium Division News, VIII, No. 3 (June, 1963), 20-22.

69. Ruehle, Arch. "The Story Behind the Story," Mallinckrodt Magazine, IX, No. 2 (February, 1964), 21-23.

70. Ruhoff, John. "The First Fifty Critical Days," as told to Pat Fain, Uranium Division News, VII, Nos. 3 & 4 (June, 1962), 3-9.

71. Ryker, Ralph, and Ziegler, Bill. "This Business of Uranium Sampling," Uranium Division News, VII, No. 5 (October, 1962), 13-16.

72. Schafer, Emmett. "Safety Plateau . . . a Good Place to Be," Uranium Division News, VI, No. 2 (April, 1961), 6-7.

73. Schlueter, Harold. "Twenty Years of Plant Security Without a Serious Violation," Uranium Division News, VII, Nos. 3 & 4 (June, 1962), 54-56.

74. Seaborg, Glenn T. Letter to Harold E. Thayer, president of Mallinckrodt Chemical Works, Uranium Division News, VII, Nos. 3 & 4 (June, 1962), inside cover.

75. Shearer, Robert. "Five Who Have Served as Managers," Uranium Division News, VII, Nos. 3 & 4 (June, 1962), 22-24.

76. Stevenson, John. "Spin Casting -- A New Technique," Mallinckrodt Magazine, X, No. 7 (July, 1965), 16-17.

77. Swaney, Dave. "New Reference Sampler at Weldon Spring," Mallinckrodt Magazine, IX, No. 4 (April, 1964), 2-5.

78. Teter, Gene. "New Acid Recovery System," Uranium Division News, V, No. 3 (June, 1960), 2-4.

79. Thayer, Harold E. "The President's Message," Uranium Division News, XI, No. 6 (June, 1966).

80. Todd, James. "Fluid Bed -- Most Important Change in Denitration System in Twenty-two Years," Mallinckrodt Magazine, IX, No. 6 (June, 1964), 12-15.

81. Toomey, Robert. "Anatomy of a Project," Uranium Division News, VI, No. 4 (December, 1960), 26-29.

82. "Top Management Changes in the Uranium Division," Uranium Division News, VI, No. 5 (October, 1961), p. 7.

83. Trask, Warren. "Major Production Link in A.E.C.," Uranium Division News, VIII, No. 3 (June, 1963), 2-6.

84. Tuthill, S., and Shepardson, J. "That Special Ingredient," Uranium Division News, VII, Nos. 3 & 4 (June, 1962), 47-53.

85. "Uranium Division Wins Three Safety Awards," Uranium Division News, XI, No. 6 (June, 1966), 4-5.

86. Vie, Jerry. "The Divided Dingot: Another Bid for More Metal Business," Uranium Division News, VIII, No. 2 (April, 1963), 14-15.

Reports

87. Alnutt, D., et al. The Preparation of Pure Uranyl Nitrate Solution from Impure Sodium Di-uranate. MCW-2. Mallinckrodt Chemical Works, January 7, 1946.

88. Anderson, T. O., Knecht, W. S., and Weber, L. G. "Uranium Metal by Bomb Reduction," Research and Development Progress Report. NYO-1352. St. Louis: Mallinckrodt Chemical Works, January 15, 1953.

89. Berry, N. E., *et al*. *Process Development Quarterly Report, Part II*. MCW-1388. An AEC Research and Development Report. St. Louis: Mallinckrodt Chemical Works, May 1, 1956.

90. Braddock, T., *et al*. *Mallinckrodt Chemical Works -- Green Salt Pilot Plant: Final Report*. NYO-5029. St. Louis: Mallinckrodt Chemical Works, March 22, 1943.

91. Buntz, B. J., *et al*. "Investigation of Crucibles for Uranium Melting," *Process Development Quarterly Report, Part II*. NYO-1363. St. Louis: Mallinckrodt Chemical Works, May 1, 1954.

92. Buntz, B. J., and Weber, L. G. "Casting of Uranium Metal," *Research and Development Progress Report*. NYO-1352. St. Louis: Mallinckrodt Chemical Works, January 15, 1953.

93. Conrad, Charles R. *The Preparation of Pure Uranyl Nitrate From Black Oxide*. MCW Research Report No. 1. St. Louis: Mallinckrodt Chemical Works, April 1, 1946.

94. Fariss, R. H. *Report on Pilot-Plant TBP-Hexane Cycle Operation*. A Report Prepared by the Mallinckrodt Chemical Works, Uranium Division. St. Louis, October, 1955.

95. Fellows, J. A. *Bomb Reduction, Forging and Extrusion of Uranium and Uranium Alloys*. *Book I of Fuel Elements Conference*. TID-7546. March, 1958.

96. Fleming, W. S., Piper, R. D., and Kennelley, J. A. *Topical Report: Electrolytic Recovery of Scrap Uranium Metal*. MCW-1494. An AEC Research and Development Report, prepared by the Uranium Division, the Mallinckrodt Chemical Works. Weldon Spring, August 19, 1966.

97. Fuel Element Development Committee. *Annual Summary Report: Mallinckrodt Chemical Works, Weldon Spring Site, July 1, 1958 to June 30, 1959*. St. Louis: Mallinckrodt Chemical Works, August, 1959.

98. Graham, James C. *Plant 4 Progress Reports*: MCW-157 (12/20/48); MCW-155 (12/6/48); MCW-153 (11/19/48); MCW-112 (9/1/48); MCW-149 (11/1/48); MCW-143 (10/15/48); MCW-139 (10/4/48); MCW-133 (9/14/48); MCW-115 (9/14/48); MCW-128 (7/26/48); MCW-104 (7/13/48). St. Louis, Mallinckrodt Chemical Works.

99. Harrington, C. D. *Preparation and Properties of Uranium Dioxide Powder*. Book II of *Fuel Elements Conference*. TID-7546. March, 1958.

100. Huston, S. H., and Simmler, J. R. *An Investigation of the Sampling Procedure of the Ore Assay Procedure*. MCW-136. September 21, 1948.

101. Huston, S. H., and Ziegler, W. A. "Evaluation of a New Falling-Stream Sampling System to be Used as a Reference at Weldon Spring," *Tenth Annual AEC-Contractor Nuclear Materials Management Meeting*.

102. Janka, Ray. "Production of Large Derbies and Ingots," *Process Development Quarterly Report, Part II*. NYO-1363. St. Louis: Mallinckrodt Chemical Works, May 1, 1954.

103. Jeske, A. A., Levitz, N. M., and Petkus, E. J. *Fluidized-Bed Process For The Production Of Uranium Tetrafluoride From Uranyl Nitrate Solution*. TID-7501. Argonne National Laboratory, Chemical Engineering Division, February, 1956.

104. Keller, Wayne H. *Purification of Phosphate Residue From Previous Digestions of Uranium Oxides for Ether Extraction*. MCW Research Report No. 14. St. Louis: Mallinckrodt Chemical Works, April 1, 1946.

105. Keller, W. H. *Partial Preliminary Report on Production of Metal of Uniform Quality*. MCW-163. St. Louis: Mallinckrodt Chemical Works, January 5, 1949.

106. Keller, W. H., *et al*. *The Effect of Time Required for the Decomposition of Uranyl Nitrate Hexahydrate on the Yield of Crude Metal Produced from the Uranium Tetrafluoride Obtained Therefrom*. A Report prepared by the Mallinckrodt Chemical Works, December 9, 1948.

107. Keller, W. H., and Olmsted, B. C., Jr. *Experimental Remelt Furnace: Report No. I, Runs 1-9*. A report prepared by the Mallinckrodt Chemical Works. St. Louis, 1949.

108. Keller, W. H., and Tuthill, S. M. *Correlation of Analytical Results With Plant Procedure During the December Program for the Production of Metal of Uniform Quality*. MCW-164. St. Louis: Mallinckrodt Chemical Works, January 21, 1949.

109. Kloepper, H. C., Jr., and Lisot, O. L. "Porosity in Uranium Metal," Research and Development Progress Report. NYO-1352. St. Louis: Mallinckrodt Chemical Works, January 15, 1953.

110. Kuhlman, Carl W. Equilibrium Ratio of Hydrogen Fluoride and Water in the Fluorination of Uranium Dioxide at High Temperatures. MCW-118. St. Louis: Mallinckrodt Chemical Works, July 15, 1948.

111. Kuhlman, Carl W. Rate of Decomposition of Uranium Trioxide on Heating. MCW-225. St. Louis: Mallinckrodt Chemical Works, October 7, 1949.

112. Kuhlman, Carl W., Jr. Fluorination of Uranium Dioxide With Hydrogen Fluoride-Water Mixture. MCW-138. St. Louis: Mallinckrodt Chemical Works, September 27, 1948.

113. Kuhlman, Carl W., Jr. A Note on the Inhibitory Effect of Hydrogen Fluoride on the Hydrogen Reduction of Uranium Trioxide. MCW-147. St. Louis: Mallinckrodt Chemical Works, October 29, 1948.

114. Kuhlman, Carl W., Jr. Reduction of Uranium Trioxide to Uranium Dioxide with Anhydrous Ammonia. MCW-217. St. Louis: Mallinckrodt Chemical Works, September 8, 1949.

115. Kuhlman, Carl W., Jr. Reduction of Uranium Trioxide to Uranium Dioxide with Hydrogen -- Reaction Rates at Various Temperatures. MCW-142. St. Louis: Mallinckrodt Chemical Works, October 12, 1948.

116. Kuhlman, C. W. Reaction Rate of the Fluorination of Uranium Dioxide at a Continuously Increasing Temperature. MCW-121. St. Louis: Mallinckrodt Chemical Works, July 19, 1948.

117. Kuhlman, C. W. Reaction Rates of the Dry Hydrofluorination of Uranium Dioxide at High Temperatures: Preliminary Report. MCW-100. St. Louis: Mallinckrodt Chemical Works, June 23, 1948.

118. Lang, G. P., and Krieg, J. T. Topical Report: The Effect of Uranium Saturation on the Extraction of Elements Other Than Uranium Into Tributyl Phosphate. MCW-1440. A Report to the 135th National Meeting of the American Chemical Society, Boston, April, 1959. St. Louis, January, 1960.

119. Leaders, W. M., et al. Process Development Quarterly Report, Part II. NYO-1360. A Report Prepared by the Mallinckrodt Chemical Works. St. Louis, January 15, 1954.

120. Leifield, R. F., and Weidman, S. W. Topical Report: Factors That Influence the Spontaneous Ignition of Magnesium Thermite Bomb Reactors. MCW-1444. A Report Prepared by the Uranium Division, Mallinckrodt Chemical Works. Weldon Spring, Missouri, April 3, 1961.

121. Lepper, C. R. "Random Access EDP for Material Accountability and Production Control," Tenth Annual AEC-Contractor Nuclear Materials Management Meeting. An Abstract.

122. Mallinckrodt Chemical Works. Abstract: Jet-Mixer Column for the: 1) Extraction of Uranium from Various Refinery Feeds, and 2) Extraction of Nitric Acid from Raffinate. NYO-1357. A Report Prepared by the Uranium Division. St. Louis: Mallinckrodt Chemical Works, May 1, 1952.

123. Mallinckrodt Chemical Works. First Semi-Annual AEC Quarry Environmental Report, 1964. A Report Prepared by the Uranium Division. Weldon Spring, Mo.

124. Mallinckrodt Chemical Works. Off-Site Environmental Monitoring Report, second quarter, 1961. A Report Prepared by the Uranium Division. Weldon Spring, 1961.

125. Mallinckrodt Chemical Works. Second Semi-Annual and Annual Summary Off-Site Environmental Monitoring Report, 1962. A Report Prepared by the Uranium Division. Weldon Spring, 1962.

126. Mallinckrodt Chemical Works, Uranium Division. Process Development Quarterly Progress Report, January-March, 1962. MCW-1472. An AEC Research and Development Report. Weldon Spring, May 1, 1962.

127. Mallinckrodt Chemical Works, Uranium Division. Second Semi-Annual AEC Quarry Environmental Report, 1962. Weldon Spring, 1962.

128. McCalpin, Nona (ed.). Process Development Quarterly Report Part II. MCW-1393. An AEC Research & Development Report. Produced by the Uranium Division, Mallinckrodt Chemical Works. St. Louis, August 1, 1956.

129. Monti, J. L., Olmsted, B. C., and Keller, W. H. The Effect of Storage Time of Uranium Dioxide on the Production of Uranium Metal from Uranium Tetrafluoride. MCW-106. St. Louis: Mallinckrodt Chemical Works, January, 1949.

130. Nelson, E. N. Topical Report: Fluid-Bed Denitration of Thorium Nitrate. MCW-1511. Weldon Spring, Mo.: Mallinckrodt Chemical Works, January 19, 1966.

131. Niedrach, L. W., and Dearing, B. E. Electrowinning of Uranium from its Oxides. "Laboratory Studies." KAPL-1761. Schenectady, New York: Knolls Atomic Power Laboratory, General Electric Company, April 30, 1957.

132. Olmsted, Bruce. Metal Production Yields of Uranium Tetrafluoride Prepared from Uranium Dioxide Produced in the Continuous Screw. MCW-173. St. Louis: Mallinckrodt Chemical Works, March 2, 1949.

133. Olmsted, Bruce C. The Use of an Electric Furnace to Produce Crude Uranium Metal. MCW-151. St. Louis: Mallinckrodt Chemical Works, November 1, 1948.

134. Opie, J. V. The Preparation of Pure Uranium Tetrafluoride By A Wet Process. MCW Research Report No. 25. St. Louis: Mallinckrodt Chemical Works, April 1, 1946.

135. Oppold, W. A. Experimental Furnace Report #2. A report prepared by the Mallinckrodt Chemical Works. St. Louis, 1949.

136. Oppold, W. A. A General Study of the Distribution of Impurities in Remelted Uranium Metal and Related Subjects. MCW Research Report No. 30. St. Louis: Mallinckrodt Chemical Works, April 1, 1946.

137. Oppold, W. A. A Preliminary Report On Hot Topping Poured Billets. MCW-113. St. Louis: Mallinckrodt Chemical Works, September 9, 1948.

138. Oppold, W. A. A Report on Changed Operating Cycles For The Hydrofluorination Furnaces. MCW-114. St. Louis: Mallinckrodt Chemical Works, September 10, 1948.

139. Oppold, W. A. A Report On Controlled Pouring Rates Of Uranium Metal To Eliminate The Shrinkage Cavity In a Cast Billet. A Report Prepared by Mallinckrodt Chemical Works. St. Louis: Mallinckrodt Chemical Works, September 24, 1948.

140. Oppold, W. A. A Report On Controlled Pouring Rates Of Uranium Metal To Eliminate The Shrinkage Cavity In A Cast Billet. A Report Prepared by Mallinckrodt Chemical Works. St. Louis, October 22, 1948.

141. Oppold, W. A. A Study of the Effect of Various Factors on the Yields Obtained in the Remelting of Uranium Metal. MCW Research Report No. 29. St. Louis: Mallinckrodt Chemical Works, April 1, 1946.

142. Oppold, William A. Remelt Furnace Pilot Plant. MCW-177. St. Louis: Mallinckrodt Chemical Works, April 7, 1949.

143. Paine, R. M., Ruehle, A. E., and Lewis, G. W., Jr. Uranium Metal By Bomb Reduction: V- The Reaction of UF_4 and Magnesium Vapor. NYO-1335. St. Louis: Mallinckrodt Chemical Works, June 20, 1952.

144. Petty, W. G., Kerr, G. E. and Weber, L. G. "Investigation Of Reduction Bomb Construction." Process Development Quarterly Report, Part II. NYO-1363. St. Louis: Mallinckrodt Chemical Works, May 1, 1954.

145. Petty, W. G., Kerr, G. E., and Weber, L. G. "Magnesium Fluoride Slag Liner Development," Process Development Quarterly Report, Part II. NYO-1363. St. Louis: Mallinckrodt Chemical Works, May 1, 1954.

146. Philoon, W. C., and Edwards, R. M., and Farris, R. H. Topical Report: Development Of The TBP-Hexane Process For Uranium Purification. MCW-1441. A Report to the 135th National Meeting of the American Chemical Society, Boston, April, 1959. St. Louis, February, 1960.

147. Piper, R. "Laboratory Studies Of Casting Uranium Metal," Research and Development Progress Report. NYO-1352. St. Louis: Mallinckrodt Chemical Works, January 15, 1953.

148. Piper, R. D. Electrolytic Reduction of Uranium Hexafluoride to Uranium Metal: Abstract. A Report Prepared by the Uranium Division, Mallinckrodt Chemical Works. Weldon Spring plant, Weldon Spring, Mo.

149. Piper, R. D. Topical Report: Laboratory Development of the Uranium Electroreduction Process. MCW-1500. Weldon Spring, Mo.: Mallinckrodt Chemical Works, July 1, 1966.

150. Piper, R., and Ruehle, A. E. Uranium Metal By Bomb Reduction: III-Slag and Liner Studies. NYO-1333. St. Louis: Mallinckrodt Chemical Works, April 3, 1952.

151. Piper, R., and Ruehle, A. E. Use of Surface Active Agents To Prevent Emulsions In Slurex Extractions. NYO-1347. St. Louis: Mallinckrodt Chemical Works, August 11, 1952.

152. Piper, R. D., and Vie, J. D. Topical Report: Methods for Tapping Molten Uranium From An Electrolytic Cell. A Report Prepared by the Uranium Division, Mallinckrodt Chemical Works. Weldon Spring plant, 1963.

153. Ritchie, C. F., and Teter, E. K. Corrosion Tests Leading to the Recommendation of Illium-R for Green Salt Reactors. MCW-249. St. Louis: Mallinckrodt Chemical Works, March 14, 1950.

154. Robinson, S. N., and Todd, J. E. Plant-Scale Fluid-Bed Denitrator. A report prepared by the Mallinckrodt Chemical Works. Weldon Spring plant, 1966.

155. Ruehle, A. E., and Smith, R. P. A Preliminary Report On a Uranium-Molybdenum Compound. MCW Research Report No. 16. St. Louis: Mallinckrodt Chemical Works, April 1, 1946.

156. Ruehle, A. E., et al. Uranium Metal By Bomb Reduction: I- Introduction; II- Physical Properties of UF_4. NYO-1332. St. Louis: Mallinckrodt Chemical Works, March 31, 1952.

157. Schmieding, E. G., and Ruehle, A. E. Progress Report: Purification of Uranium Using Tributyl Phosphate as the Extractant. NYO-1319. St. Louis: Mallinckrodt Chemical Works, March 31, 1951.

158. Schmieding, E. G., and Ruehle, A. E., and Kloepper, H. C., Jr. Progress Report: Purification of Uranium Using Tributyl Phosphate as the Extractant. NYO-1325. St. Louis: Mallinckrodt Chemical Works, October 22, 1951.

159. Schroeder, C. D. Progress Report of Brown Oxide Production by Continuous Method. MCW-207. St. Louis: Mallinckrodt Chemical Works, May 26, 1949, (issued August 10, 1949).

160. Schroeder, C. D. Progress Report of Uranium Dioxide by Continuous Method, (Period 5/17/49 to 7/13/49). MCW-229. St. Louis: Mallinckrodt Chemical Works, August 2, 1949.

161. Shearer, R. W. The Reduction of Uranium Tetrafluoride With Magnesium: II- The Influence of Physical and Chemical Properties of the Magnesium on the Reaction. MCW Research Report No. 27. St. Louis: Mallinckrodt Chemical Works, April 1, 1946.

162. Shearer, R. W. Reduction of Uranium Trioxide To The Dioxide With Various Gases. A report prepared by Mallinckrodt Chemical Works. St. Louis, April, 1946.

163. Shelley, William. Preliminary Studies of a Continuous Process For The Thermal Decomposition of Uranyl Nitrate Hexahydrate. NYO-1314. St. Louis: Mallinckrodt Chemical Works, December 7, 1950.

164. Shelley, W. J., and Kuehn, M. N. *Reprint from "Economic Criteria Applied to Nuclear Materials Management."* A Report to the International Atomic Energy Agency, in Vienna, 1966. Austria: By the IAEA, 1966.

165. Shepardson, J. U., and Nelson, J. A. (ed.) *Process Development Quarterly Report*. MCW-1433. St. Louis: Mallinckrodt Chemical Works, November 2, 1959.

166. Simecek, S., and Trask, W. T. *Topical Report: The Integrated Fluid-Bed System*. MCW-1478. A Report Prepared by the Uranium Division, Mallinckrodt Chemical Works. Weldon Spring, 1963.

167. Stevenson, J. W., Buckheim, O. J., and Ruehle, A. E. "The Recovery of Uranium and HF from MgF_2 Bomb Slag." *Research and Development Progress Report*. NYO-1352. St. Louis: Mallinckrodt Chemical Works, January 15, 1953.

168. Stevenson, J. W., and Ruehle, A. E. *Uranium Metal By Bomb Reduction: VI- Ignition Temperatures of Magnesium Metal With UF_4, UO_2F_2, UO_2, U_3O_8, and UO_3*. NYO-1339. St. Louis: Mallinckrodt Chemical Works, January 31, 1953.

169. Teter, E. K. *Report of Pilot Plant Work on UF_4 Production By Fluidization*. MCW-201. St. Louis: Mallinckrodt Chemical Works, July 15, 1949 (issued July 22, 1949).

170. Teter, E. K. *Report of Pilot Plant Work on UF_4 Production By* Means of Stirred Fluidization. MCW-238. St. Louis: Mallinckrodt Chemical Works, November 15, 1949 (issued December 9, 1949).

171. Teter, E. K. *A Series of Progress Reports of Pilot Plant Work on UF_4 Production by Continuous Methods*: MCW-109 (period 7/9/48 to 8/14/48); MCW-152 (period 8/15/48 to 10/15/48); MCW-165 (issued 1/31/49); MCW-179 (issued 4/11/49); MCW-184 (period 3/16/49 to 4/15/49); MCW-205 (period 5/21/49 to 6/18/49); MCW-209 (period 6/19/49 to 7/13/49); MCW-220 (period 7/14/49 to 8/16/49); MCW-228 (period 8/17/49 to 9/24/49); MCW-243 (period 9/25/49 to 11/30/49); MCW-246 (period 11/20/49 to 1/5/50); MCW-253 (period 1/6/50 to 3/5/50); NYO-1304 (period 3/6/50 to 4/21/50); NYO-1313 (period 4/22/50 to 7/31/50); NYO-1316 (issued 12/18/50). Reports Prepared by the Mallinckrodt Chemical Works.

172. *Tubealloy Process Report*. MCW-51. St. Louis: Mallinckrodt Chemical Works, January 3, 1944.

173. U. S. Atomic Energy Commission. Expansion Program at St. Louis Area: Project History and Completion Report. TID-5886. A Report Prepared by the St. Louis Area Office. Oak Ridge, Tennessee: USAEC Office of Technical Information Extension, October, 1960.

174. U. S. Atomic Energy Commission. Mallinckrodt Chemical Works, Plant 6E: Occupational Exposure to Airborne Contaminants. MCW-18. A Report Prepared by the Health and Safety Laboratory, New York Operations Office. New York: New York Operations Office, February 5, 1954.

175. Walden, C. H. MCW (Mallinckrodt) SF Accountability Program, a Progress Report. NYO-1317. St. Louis: Mallinckrodt Chemical Works, January 2, 1951.

176. Weber, Walter, and Yeager, Harold. A Pilot Plant Study Of Removal Of Boron From Uranyl Nitrate Solutions By Ether Extraction. MCW-198. St. Louis: Mallinckrodt Chemical Works, July 22, 1949.

177. Yeager, Harold. Conditions for the Production of Chemical 264 Purified. NYO-5080. Mallinckrodt Chemical Works, February 8, 1945.

178. Yeager, Harold. The Preparation Of Uranium Trioxide From Uranyl Nitrate Solution. A Report Prepared by the Mallinckrodt Chemical Works. St. Louis, April, 1946.

179. Yeager, J. H. Monthly Progress Report of Continuous Talcum Process, Period of 5/1/48 to 5/31/48. MCW-101. St. Louis: Mallinckrodt Chemical Works, July 2, 1948.

180. Yeager, J. H. Monthly Progress Reports of Pilot Plant Work on UF_4 Production by Continuous Methods: MCW-7 (period 6/1/48 to 6/30/48); MCW-119 (period 6/1/48 to 6/30/48). Reports Prepared by the Mallinckrodt Chemical Works. St. Louis, 1948.

181. Ziegler, W. A., et al. An Evaluation of the Weldon Spring Feed Preparation and Sampling Plant. MCW-1422. St. Charles, Mo.: Uranium Division, Mallinckrodt Chemical Works, December 1, 1958.

Unpublished Material

182. Anonsen, S. H. "Tube Alloy (Uranium) Health Protection." An unpublished report of all health procedures followed at Mallinckrodt Chemical Works' Uranium Division from the beginning of their involvement in the Manhattan Project. December, 1944.

183. Breslin, Alfred J., and Glauberman, Harold, and Loysen, Peter. "Mallinckrodt Chemical Works - Plant 7: Occupational Exposure to Radioactive Dust." Unpublished Report Prepared by the Health and Safety Division, New York Operations Office, December 22, 1952.

184. Davidson, Ward F. "Nuclear Energy for Power Production." Paper submitted for presentation at the Sectional Meeting of the World Power Conference, The Hague, September 2-9, 1947. (Mimeographed.)

185. Edwards, R. M., and Hyman, E. D., and Stevenson, J. W. "Uranium Tetrafluoride Plant." Mallinckrodt Chemical Works, St. Louis, December, 1955.

186. Harrington, Charles D. "The Commercial Production of Uranium Metal." Unpublished report. St. Louis: Mallinckrodt Chemical Works.

187. Harrington, Dr. C. D. "Technical Division Activities for 1955." St. Louis: Mallinckrodt Chemical Works, December 27, 1955.

188. Kelley, W. M. "History of the M.C.W. Project Plants, 1947-1950; part II." Unpublished, rough-draft report. St. Louis, August, 1950.

189. Kelley, W. M. "History of the M.C.W. Project Plants, 1947-1950; part III." Unpublished, rough-draft report. St. Louis, August, 1950.

190. Kelley, W. M. "History of the M.C.W. Project Plants, 1947-1950; part IV-d." Unpublished, rough-draft report. St. Louis, August, 1950.

191. Kornfeld, Robert. "Indoctrination Report." A brief, unpublished report on the organization of Mallinckrodt Chemical Works' Uranium Division. July 28, 1958.

192. Mallinckrodt Chemical Works. "The Atomic Adventure." Chapter VII of 1967 draft of official history of Mallinckrodt Chemical Works. Prepared under the direction of Dr. Paul A. Krueger, Mallinckrodt Chemical Works, St. Louis.

193. Mallinckrodt Chemical Works. "Uranium Division Emergency Manual." October, 1964.

194. Mallinckrodt Chemical Works, Uranium Division. "AEC Health Protection Study Committee Audit at Weldon Spring and St. Louis, Missouri." Compilation of documents, memoranda, and data taken from the files of the Health Department, Mallinckrodt Uranium Division. August 19-20, 1957.

195. Mallinckrodt Chemical Works, Uranium Division, and the Independent Union of Atomic Workers. "An Agreement between M.C.W. Uranium Division and the IUAW, effective August 19, 1963."

196. Mallinckrodt Chemical Works, Uranium Division. "Criticality Emergency Manual." Unpublished manual for Uranium Division employees. Weldon Spring, September, 1964.

197. Mallinckrodt Chemical Works, Uranium Division. "Organization Manual." June, 1955.

198. Mallinckrodt Chemical Works, Uranium Division. "The Atomic Energy Commission -- Weldon Spring Plant: Mallinckrodt Operation." Unpublished speech given before the Chambers of Commerce of certain St. Louis County townships. St. Louis, December, 1955.

199. Mallinckrodt Chemical Works, Uranium Division. "Tour Guide Book: Feed Materials Plant." Weldon Spring, Missouri, February, 1962.

200. Mallinckrodt Chemical Works, Uranium Division. "Uranium Division Emergency Plan, Weldon Spring Plant." Policy, plans and procedures to be followed in event of an emergency. Revised, March, 1962.

201. Mallinckrodt Chemical Works, Uranium Division. "Weldon Spring, An Introduction." A printed brochure on the Weldon Spring Plant.

202. Mallinckrodt Chemical Works, Uranium Division. "Standard Operating Procedures for Metal Plant - 301 Area, Weldon Spring." General operating procedures, special instructions, and health and safety regulations for each operation. May 17, 1961.

203. Mallinckrodt Chemical Works, Uranium Division. "Standard Practice Manual." Vol. I, II, III. A collection of Standard Practice Instructions, up to May, 1966.

204. Mallinckrodt Chemical Works, Uranium Division. "Summary of Health Protection Practices." Revised. Unpublished document prepared by the Health and Safety Department, summarizing protection functions carried out at the Weldon Spring Feed Material Processing Plant. Weldon Spring, 1965.

205. Mallinckrodt Chemical Works, Uranium Division Criticality Committee. (ed.) "M.C.W. Manual for Criticality Safeguards in Processing 0.95 W/O Enriched Uranium." Weldon Spring, November, 1962.

206. Mallinckrodt Chemical Works, Uranium Division Criticality Committee. (ed.) "M.C.W. Manual for Criticality Safeguards in Processing 1.5 W/O Enriched Uranium." Weldon Spring, September, 1963.

207. Opie, John V. "Instrumentation for Process Control." Unpublished report. St. Louis: Mallinckrodt Chemical Works, January 31, 1967.

208. Petty, W. G. "Dingot Report for the Reactor Handbook." Unpublished report. St. Louis: Mallinckrodt Chemical Works, Uranium Division, February 4, 1956.

209. Robertson, W. J. "Thorium Operations at MCW." Unpublished report. St. Louis: Mallinckrodt Chemical Works, March 2, 1967.

210. Robinson, S. N. "Indoctrination Report," Unpublished report on the personnel, organization, functions and activities of most of the departments within the Uranium Division, Mallinckrodt Chemical Works, October 31, 1956.

211. Ruehle, A. E. "Trends in Uranium Processing." Paper presented at the Sixty-third National Western Mining and Energy Conference, Denver, April 23, 1960. (Mimeographed.)

212. Ruhoff, John R. "Problem: 42-17 Uranium Oxide S. L.: Tube Alloy - Experimental." Laboratory report, made at the Uranium Division, Mallinckrodt Chemical Works, St. Louis, May 4-8, 1942. (Typed.)

213. Shelley, W. J. "Mallinckrodt Weldon Spring Safety Experience." Unpublished report to employees of the safety record at Weldon Spring from 1963 through 1966. December, 1966.

214. Shelley, William J. "Uranium Division Management and Organization History." Drafts 2/24/67 and 2/28/67. St. Louis: Mallinckrodt Chemical Works. (Mimeographed.)

215. Thayer, Harold E. "From a Tradition of Integrity, the Heritage of Growth." A speech given before the Newcomen Society in North America. St. Louis: Mallinckrodt Chemical Works, February, 1967. (Printed.)

216. Thayer, Harold E. "Weldon Spring Story." Talk given by Thayer, June 5, 1956. (Typewritten.)

217. United States Atomic Energy Commission. "AEC Will Consolidate Feed Materials Operations." Press Release. St. Louis, April 28, 1966.

218. United States Atomic Energy Commission. "U. S. Presents Special Sets of Books on Peaceful Uses of Atomic Energy to Officials at Third Geneva Conference." Press release, September 2, 1964.

219. Ziegler, W. A. "Sampling, Sample Preparation, and Analytical Methods Development." Unpublished report. St. Louis: Mallinckrodt Chemical Works, February 28, 1967.

Other Sources

220. Mallinckrodt Chemical Works. Personal interview with Mont G. Mason, Co-ordinator, air and water pollution control. January 8, 1967.

221. Mallinckrodt Chemical Works, Uranium Division. Correspondence with Jerry D. Vie. 1966-1967.

222. Mallinckrodt Chemical Works, Uranium Division. Personal interviews and correspondence with Edward Monaco. 1966-1967.

223. Mallinckrodt Chemical Works, Uranium Division. Personal interviews and correspondence with Archie E. Ruehle. 1966-1967.

224. Mallinckrodt Chemical Works, Uranium Division. Personal interviews and correspondence with William J. Shelley. 1966-1967.

225. Mallinckrodt Chemical Works, Uranium Division. Personal interviews with John V. Opie. 1966-1967.

226. Mallinckrodt Chemical Works, Uranium Division. Personal interviews with John W. Stevenson. 1966-1967.

227. Mallinckrodt Chemical Works, Uranium Division. Personal interviews with Wilbert J. Robertson. 1966-1967.

228. Mallinckrodt Chemical Works, Uranium Division. Personal interviews with Wiley J. S. Smith. 1966-1967.