UNCLASSIFIED

CT-2958

E-1

45 P.

This document is
PUBLICLY RELEASABLE

Hugh Kinsey
Authorizing Official
Date: *9-7-11*

Iowa State College

Contract No. W-7405, eng-82
W-7405, eng-7
OEMsr 433
OEMsr 410

Volume 12-B

EARLY METHODS FOR CASTING URANIUM AT IOWA STATE COLLEGE

by

C. F. Gray

July 1945

CLASSIFICATION CANCELLED

DATE FEB 18 1957 *jet*

For The Atomic Energy Commission

H. R. Canale
Chief, Declassification Branch

[Redacted]
Office of Technical Services
Department of Energy
Washington 25, D.C.

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, express or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission to the extent that such employee or contractor prepares, handles or distributes, or provides access to, any information pursuant to his employment or contract with the Commission.

UNCLASSIFIED

DECLASSIFIED

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

~~CONFIDENTIAL~~

EARLY METHODS FOR CASTING URANIUM AT IOWA STATE COLLEGE*

Work done by F. H. Spedding, H. A. Wilhelm, and C. F. Gray

Written by: C. F. Gray

Uranium production in the United States, at the beginning of the work at Ames in February 1942, was carried out almost entirely by two small scale production plants. One was Metal Hydrides located at Beverly, Massachusetts, and the other, Westinghouse Lamp Company of Bloomfield, New Jersey. At Metal Hydrides, the uranium oxide was reduced with calcium hydride in a steel retort heated to 960°C . The resulting mass, after cooling under vacuum, was crushed and leached with 2 per cent acetic acid to give a wet powdered metallic uranium. This was dried in a low-temperature oven through which circulated a stream of dry nitrogen. The dry powder was very pyrophoric and was handled under a carbon dioxide atmosphere at -15°C while being pressed into briquets of the desired form. These briquets were then sintered to give metallic uranium which was of low density and purity.

At Westinghouse, KUF_5 or UF_4 was electrolyzed in a hot $\text{CaCl}_2\text{-NaCl}$ bath (80-20) to give a deposit of finely divided uranium metal. The hot electrolyte was held in a graphite crucible at a temperature of 900°C . The graphite crucible served as the anode and a molybdenum bar was used as the cathode. At the end of electrolysis the cathode was withdrawn and smothered in dry sodium chloride. When cool the deposit on the cathode was broken loose with an air hammer; then crushed, ground, and leached to free the finely divided metal. This was then rinsed with distilled water, washed in alcohol, and then in ether and finally dried in a vacuum oven.

*Based on CC-148, CC-176, CC-177, CC-178, CC-205, CC-238, CC-253, CC-298, CT-393, CT-422, CT-490, CT-542, CT-609.

~~CONFIDENTIAL~~

This metal was less pyrophoric than that prepared by Metal Hydrides and was stored temporarily in the form of pressed briquets under a carbon dioxide atmosphere. By means of an Ecco oscillator, Westinghouse fused the pressed uranium by self induction in beryllia crucibles and flowed it into beryllia molds of the desired shapes. This operation was carried out under high vacuum. The final product was of somewhat better purity and had a much higher density than the metal produced at Metal Hydrides.

One serious disadvantage to the Westinghouse process, however, was the dependence on beryllia ware. This material, although satisfactory for casting small laboratory amounts, was not satisfactory for large scale production casting. The main objections to beryllia crucibles and molds were (1) the high cost of beryllia of the required purity, (2) the fragileness of large crucibles and molds, (3) the difficulties of fabrication, and (4) the actual shortage of enough beryllia to meet the demands of the proposed uranium production. In view of the above problems and the probability that the free uranium, however reduced, must be cast into its final form or some easily convertible intermediate form, the Iowa State College Laboratory started a search for a new material in which uranium could be melted and cast.

Early in June 1942, it was decided to experiment with graphite as a crucible and mold material, especially since it was easily fabricated and a self inductor. This was done in spite of the fact that the high melting point of uranium (<1800°C as reported in the handbooks at that early date) pointed to the possibility of reaction difficulties in the use of graphite in contact with liquid uranium. The first test⁽¹⁾ with graphite was set up as shown in Figure 1. A rectangular block of uranium (11) (370 g. of

3

DECLASSIFIED

Fig. 1. First successful melting of uranium in graphite. (1) Glass viewing window, (2) water connection, (3) vacuum connection, (4) rubber gasket, (5) water-cooled vacuum head, (6) closed-end quartz tube, (7) Norblack insulation, (8) graphite viewing chimney, (9) induction coil, (10) graphite crucible, (11) uranium block, (12) K-30 insulation brick, (13) induction coil support.

DECLASSIFIED

metal) was placed in a graphite crucible (10) having a hemispherical bottom. This crucible was packed in Norblack insulation (7), a finely divided amorphous carbon. The system was evacuated through opening (3) to about 500 microns and the crucible heated by the induction coil (9) connected to an Ajax mercury arc converter. The temperature was measured with an optical pyrometer by focusing on the uranium block through the viewing glass (1) and the graphite viewing chimney (8). The temperature of the metal was raised to 1650°C and then allowed to cool to room temperature. Unpacking the crucible revealed that the uranium had melted and filled the hemispherical crucible bottom, and that it easily fell out of the crucible when inverted. A skin seemed to have formed on the outside of the original block and part of this skin retained its rectangular shape to some extent and projected as a thin shell above the casting. No reaction with the graphite was detected at this time; however, later observations showed that a very thin uranium carbide layer was formed where the metal came in contact with graphite at such temperatures.

This lower temperature, 1650°C , for melting was encouraging and the promising results obtained from the first experiment prompted further investigations (1)(3) on a means of separating the skin from the molten metal and producing sound castings. As a result, the crucible and mold combination shown in Figure 2 was designed. A uranium metal block (4) was placed in the graphite crucible (3) and on the graphite grill (5) which contained long slots as shown in section XX. The crucible was packed in the same manner as the one shown in Figure 1 and the same heating procedure was followed. The results were very satisfactory in that about

5

Fig. 2. First successful drip casting of uranium in graphite. (1) Viewing chimney, (2) graphite lid, (3) graphite crucible, (4) uranium block, (5) grill, (6) graphite mold. (Cross section of uranium block is not shown in section X-X).

DECLASSIFIED

70 per cent of the uranium block melted and ran through the grill to give a bright, clean, and well-formed hemispherical casting in the mold (6). The skin remained on top of the grill. Both crucible and mold were heated and under these conditions the mold was referred to as a "hot mold". This method of melting and flowing the molten metal through a grill was often referred to as "drip casting". With the above results, it was definitely concluded that uranium could then be successfully melted in graphite crucibles and cast into graphite molds.

The success of the above experiments brought a number of requests for uranium to be cast into various shapes. Sketches of some of the crucible-mold units used for these special castings are shown in Figure 3, 4, 5, 6, 7, 8, 9, 10 and 11. All the melting and casting units shown in these figures were packed in a manner similar to the one shown in Figure 1, although the insulation, the size of the equipment, the heating procedure, and other minor points were varied somewhat from time to time. The effects of these variations will be discussed later. Although the very early castings were made with uranium obtained from Westinghouse or Metal Hydrides, a large part of the work described here was carried out with metal produced by the Iowa State College process in which calcium⁽⁴⁾ or magnesium⁽⁵⁾ was used to reduce uranium tetrafluoride in a closed bomb.

The crucible and mold shown in Figure 3 were used to cast a 4 in. hemisphere⁽³⁾⁽⁶⁾⁽⁷⁾. The hemisphere had sharp detail and was solid except for a small pipe which formed on the top side. This brought out the necessity of a special cooling procedure to eliminate voids in the tops of the castings. The crucible-mold assembly shown in Figure 4 was used to cast a series of bars $\frac{1}{2} \times \frac{1}{2} \times 2$ in.⁽⁸⁾⁽⁹⁾⁽¹⁰⁾. This assembly was successful and no difficulties with pipes were experienced.

Fig. 3. Graphite crucible-mold for drip casting 4 in. hemisphere. (1) Viewing chimney, (2) graphite lid, (3) graphite crucible, (4) graphite grill, (5) graphite mold.

DECLASSIFIED

Fig. 4. Graphite crucible-mold for casting $1/2 \times 1/2 \times 2$ in. bars.
 (1) Viewing chimney, (2) graphite lid, (3) graphite crucible, (4) graphite mold, (5) bottom cap.

DECLASSIFIED

At this time some experimental runs were made with static atmospheres of hydrogen and argon gases⁽²⁾. In the case of the hydrogen, a large volume of extremely pyrophoric black powder was obtained. A suspicion that this powder was uranium hydride was later confirmed by the Iowa State College Laboratory⁽¹¹⁾. As a furnace atmosphere for casting, hydrogen was definitely unsatisfactory under the conditions used. One casting was attempted in a graphite crucible under air at atmospheric conditions⁽¹²⁾. This crucible was directly open to the air through a 1/4 in. I.D. viewing chimney. A good yield, about 80 per cent, was unexpectedly obtained. Experiments with argon atmospheres showed that fair castings could be made in that gas; however, much better results were obtained in a vacuum and the work with various gaseous atmospheres was discontinued.

While measuring the temperature with an optical pyrometer in some of the above experimental castings, the uranium blocks were actually observed to melt and flow at temperatures ranging from 1080°C to 1150°C⁽³⁾. This temperature range was far lower than the melting point value of <1800°C which was given in the current literature. The high values for the melting point appearing in the literature at the time can probably be explained by the formation of a tenacious skin on the uranium blocks during the preheating stage. This skin was found to be very troublesome, especially for casting small quantities where a block of metal would probably have to be heated to a much higher temperature before the skin would be sufficiently weak to break under the weight of the molten uranium. In the case of a larger block of free uranium, the pressure on the skin was consequently larger and the breaking of the skin occurred at lower temperatures as indicated above. The low melting temperatures were observed only for large blocks of cast uranium.

For unmelted finely divided uranium such as sintered blocks from Metal Hydrides very high temperatures (1600 - 1800°C)⁽³⁾ were necessary to make it flow. On recasting, however, Metal Hydrides metal flowed in the lower temperature range.

After massive uranium had been successfully produced by the reaction of uranium tetrafluoride and calcium⁽⁴⁾ at Iowa State College, the request was received to cast the massive metal into the form of cylinders about 2.25 in. in diameter. Various sizes of these cylinders were requested varying in weights from 4.5 to 7 lbs. They were referred to as "eggs" and the calcium reduced lumps of crude massive metal were termed "biscuits" in all production operations.

All crucibles and molds⁽¹³⁾⁽¹⁴⁾⁽¹⁵⁾⁽¹⁶⁾⁽¹⁷⁾ were made of graphite and were designed to cast cylinders sufficiently long that each could be cut into two or possibly three eggs. Some of the crucible-mold combinations used for casting eggs are shown in Figures 5, 6, 7, 8, 9, 10 and 11. The single unit for melting and casting shown in Figure 5 was satisfactory for small amounts, but very wasteful of graphite. When cylinders were cast into a hot mold, the metal formed a thin carbide layer on the graphite surface which bonded the uranium casting to the graphite mold. For this reason the mold shown in Figure 5 was lost with each run. To eliminate the loss of graphite the crucible-mold unit shown in Figure 6 was designed with a graphite liner in the mold cavity. In this model the liner had to be precision fitted to the mold so that metal could not flow between the mold wall and liner. In this work the fabrication exceeded the cost of graphite saved, and consequently the mold liner was discontinued. The pins (4) used to hold down the grill in Figure 5 proved unnecessary in setups where the crucible (3)

Fig. 5. First graphite crucible mold for casting eggs. (1) Viewing chimney, (2) graphite lid, (3) graphite crucible, (4) graphite pin, (5) graphite grill, (6) graphite mold.

NOT REPRODUCED

Fig. 6. Revised graphite egg casting unit. (1) Viewing chimney, (2) lid, (3) crucible, (4) grill, (5) mold, (6) mold sleeve.

DECLASSIFIED

and mold (6) consisted of one piece of graphite.

The crucible shown in Figure 7 was designed for casting with the mold partially below the heating zone of the induction coil. It was hoped that the ingot from a partially heated mold might be removed from the mold without breaking it. Tests showed that drip castings made into molds that were partly outside the heating zone had folds at the bottoms, or cold ends of the molds and that the castings stuck to the graphite at the top, or hot ends of the molds. This latter condition prevented the re-use of the graphite molds.

Early in the development of the reduction process, a sleeve above the grill, as shown in Figure 7, was added to prevent the slag and the large excess of calcium in the biscuits from reacting with the side of the crucible. Tests showed that this sleeve was effective in protecting the sides of the crucible so that the grill could be removed after use, but little was gained since the bottom mold was lost. These difficulties led to the design shown in Figure 8. Here, the crucible and mold were made into two separate parts. The mold was heated above the melting point of uranium to eliminate the brainy structure obtained by drip casting into a cold mold and consequently the mold was lost with each casting. The crucible (3) had a removable grill (6) which was held in place by graphite pins(5). With this arrangement the crucible and grill could be used more than once.

To meet production demands the casting assembly shown in Figure 9 was developed. This model had a three-hole mold and could yield up to 6 or 9 eggs per good casting. A separate set of openings was provided for each mold hole in the grill. Both the crucible and the grill rested on the mold. One disadvantage in this design was that the molds were often only partially filled.

DECLASSIFIED

Fig. 7. Revised graphite egg casting unit with protection sleeve for crucible. (1) Viewing chimney, (2) lid, (3) crucible, (4) crucible protection sleeve, (5) grill, (6) mold.

Fig. 8. First two-piece graphite egg casting unit. (1) Viewing chimney, (2) lid, (3) crucible, (4) crucible protection sleeve, (5) pin, (6) grill, (7) mold.

DECLASSIFIED

Fig. 9. First multiple mold graphite egg casting unit. (1) Viewing chimney, (2) lid, (3) crucible, (4) crucible protection sleeve, (5) grill, (6) mold.

DECLASSIFIED

As a result, fewer eggs were obtained than if two molds had been filled and the third left empty or nearly so. To correct this an annular spillway was cut into the tops of the molds to connect all molds together and the grill was slotted for only two of the molds. This revised model is shown in Figure 10. As a result, the two molds with grills filled and then the excess ran into the third mold. The importance of the above spillway was reduced at about this period by improvements in the casting procedures and in the quality of the crude. Since the casting yields had become moderately consistent, the correct weight of charge could be calculated to give approximately three full molds from each run.

After the calcium reduction of uranium tetrafluoride had been studied more carefully it was possible to produce crude uranium biscuits that contained only small amounts of slag and calcium. As a result of these improvements, there was much less attack on the crucible walls during melting and the use of the crucible sleeve became unnecessary. Further, it was discovered at this time that if a crucible was unpacked while quite hot the slag remaining in it would ignite and burn to form a black powder which was referred to as "dross". After some experiments⁽¹⁷⁾ with heating a used crucible with a gas flame it was found that all the residue remaining from casting could be burned out. This procedure was put into practice and all crucibles were re-used until defects developed and the crucibles were considered unsafe for further use.

With the improvements in cleaning the crucibles by burning out the residue and the reduction in the large excesses of slag from biscuits reduced with calcium, the casting assembly shown in Figure 11 was designed and put into operation. In this model the grill (4) was only a small

Fig. 10. Revised multiple mold graphite egg casting unit. (1) Viewing chimney, (2) lid, (3) crucible, (4) crucible protection sleeve, (5) grill, (6) circular spillway, (7) mold.

DECLASSIFIED

Fig. 11. Graphite crucible and quadruple mold unit. (1) Viewing chimney, (2) lid, (3) crucible, (4) grill, (5) mold.

removable disc in the center of the crucible (3). The multiple mold (5) contained four holes and the molten metal dripped into the center where it was equally carried to all four mold cavities by a spillway cut into the top of the mold. If one cavity did fill before another one, metal from the full one flowed into the adjacent cavities.

In all the above work, the grill had consisted of a series of long slots varying in width (with different grills) from $1/8$ to $3/8$ in. The main purpose of the grill was to act as an agent in breaking and retaining the skin while the metal flowed into the mold. To break the skin, it was desirable to have a large grill opening, yet not so large that pieces of skin would be carried into the mold. In the case of a very heavy skin, which was occasionally encountered in recasting under a poor vacuum, grill slots $1/4$ in. wide were not wide enough. In such cases a small ring (Figure 12) was set on the grill and the metal was placed on top of this ring. The ring, about 2 in. in diameter, had its points of support for the skin far enough apart to cause the molten metal to break the skin. Such heavy skins were only encountered while Norblack was used as insulation. Norblack, a finely divided carbon, probably absorbed large quantities of gases and liberated them under the vacuum at high temperatures to produce the heavy skins on uranium.

After a few experiments with various types of grills a very simple one⁽¹⁴⁾ was developed for the drip casting into hot molds. It is shown in Figure 13. In this model no spillway was cut between the holes in the mold. A hole was cut in the bottom of the crucible large enough to overlap the mold holes about $1/2$ in. at the widest point. This was satisfactory and very easy to fabricate.

DECLASSIFIED

Fig. 12. Skin breaking ring.

Fig. 13. Simplified graphite egg casting unit. (1) Viewing chimney, (2) lid, (3) crucible, (4) mold.

Norblack insulation, the first type used, had proved very difficult to handle in large systems in a vacuum. Its resistance to air flow was so high that it seemed to hold air and then explode in a vacuum even when the vacuum was produced slowly. Small explosions or puffs were also observed in Norblack at high temperatures, after it had been successfully evacuated. This might be explained by large volumes of adsorbed gases being given off over a short temperature range during the rapid induction heating. With Norblack a vacuum could be obtained only very, very slowly and then with no certainty that an explosion would not result and fill the vacuum line, filter, and pump with fine carbon. The best way found to use Norblack in a vacuum is shown in Figure 14. The crucible-mold (8)(9) and Norblack (6) were packed in a large porous container (5) constructed from K-30 insulation brick. This unit was then placed in a closed-end quartz tube (4) on blocks of K-30 insulation brick (10) and the system evacuated in the usual manner through opening (2). In this case, air did not travel more than one inch through Norblack at any place; while without the porous container, air had to travel at least 18 in. in some places.

The discovery of a lower melting point for uranium as described above made lower maximum heating temperatures possible, and subsequently the abandonment of Norblack insulation. Little trouble was experienced in breaking the skins after Norblack insulation was abandoned. The strong skins obtained when using Norblack can be explained by assuming that large volumes of gases are released from this finely divided carbon under vacuum at high temperatures to react with the uranium during heating. For insulation at the lower temperatures employed, coarsely crushed K-30 insulation brick with the fine dust removed was satisfactory. It was used throughout

DECLASSIFIED

Fig. 14. Successful casting assembly for using Norblack insulation in a vacuum. (1) Viewing window, (2) vacuum connection, (3) water-cooled vacuum head, (4) closed-end quartz tube, (5) K-30 insulation brick container, (6) Norblack insulation, (7) viewing chimney, (8) crucible, (9) mold, (10) K-30 insulation brick base.

DECLASSIFIED

the drip casting period except for a short interval during which, due to the shortage of K-30 insulation brick, it was necessary to try some coarse graphite chips from the fabrication of crucibles and molds. This was satisfactory, though inferior to the crushed K-30 insulation brick.

The vacuum for the casting assemblies described above was produced by either a Cenco Megavac or Hypervac 20 pump. A sketch of all the vacuum system except the vacuum pump is shown in Figure 15. The water-cooled head (6) on the closed-end quartz tube (7) was connected by approximately 6 ft. of 1/2 in. I.D. rubber hose (18) to a dust trap filled with glass wool (15) which was moistened with vacuum pump oil. The glass wool dust trap (15) was about 2 in. in diameter and 10 in. long and was connected to the vacuum pump at (17) by about 2 ft. of 1/2 in. I.D. rubber hose (not shown). A small mercury manometer (14) was sealed to the glass tube leading to the dust trap. A small stopcock (16) was also sealed to this glass tube for releasing the vacuum. Pressures of less than 2 mm. were obtained with above apparatus.

As pointed out earlier, the temperatures were measured with an optical pyrometer by sighting on the uranium biscuit through the viewing window and the graphite viewing chimney. This method was satisfactory for recast metal, but very unsatisfactory for determining the temperature of biscuits produced with a large excess of calcium. For such biscuits there were small explosions or ebullitions of vapors at about 1350° to 1375°C (maximum temperature 1400°C). In the case of biscuits reduced with a large excess of magnesium there were also sudden ebullitions of vapors at about the melting point of uranium. The smoke produced by these explosions and ebullitions of vapors filled the viewing chimney and adhered to the viewing glass, making accurate measurements impossible.

REF ID: A50177

Fig. 15. Complete drip casting unit. (1) Water connection, (2) viewing glass, (3) water connection, (4) vacuum connection, (5) gasket, (6) water-cooled head, (7) quartz tube, (8) viewing chimney, (9) induction coil, (10) insulation, (11) crucible, (12) mold, (13) K-30 insulation brick base, (14) mercury manometer, (15) glass wool dust trap, (16) stop cock, (17) vacuum connection to forepump, (18) rubber hose.

DECLASSIFIED

The only changes made in the heating procedure during the above development were to change the rate of heating, the maximum temperature, and the time held at the maximum temperature. Changes in the rate of heating showed no marked effects in any direction except faster rates made the ebullition of vapors from high calcium or magnesium crude metal at higher temperatures more violent. The first casting was made with a maximum temperature of 1650°C . After the melting point was proven to be close to 1100°C the maximum temperature was lowered in progressive steps to 1200°C at which point some large amounts of impurities (N, Ca, Mg) were detected. Further experiments at various temperatures showed that heating the uranium in the molten state under a vacuum eliminated certain impurities and increased the carbon content. A heating procedure developed at this time was approximately as follows:

1. With the charge of uranium biscuits placed in the upper portion of the induction coil and the bottom of the mold approximately even with the bottom of the induction coil, heat to about 1300°C .
2. Lower the rate of heating and maintain the temperature at 1300°C for about 10 min.

Many changes were made in the cooling procedure which became of prime importance in producing castings with good surfaces and no pipes. Egg specifications permitted the shipment of only those eggs free of pipes and with surface pits not greater than $1/16$ in. in depth. Without special cooling procedures, cylinders cast long enough to be cut into three eggs would have one or more pipes running the greater part of their length with the net result that probably only one satisfactory egg per cylinder would be obtained. To eliminate these conditions, cooling must progress from the bottom to the top of the mold so that molten metal will be available to fill

the pipe as it tends to form. This condition was approached by applying heat with the induction coil to the top of the mold as the bottom cooled. While the top of the mold was being heated its position with reference to the coil was changed by lowering the quartz tube assembly shown in Figure 15 with respect to the coil. Supports of various heights were provided to give adjustment in any position. A typical cooling procedure was as follows:

1. With the crucible-mold assembly in the heating position as described above (temperature of both crucible and mold above melting point of uranium), lower the crucible to 2 in. below the bottom of the induction coil and heat slowly while the crucible temperature falls to 1175°C .
2. Lower the crucible-mold assembly $2\frac{1}{2}$ in. more or $4\frac{1}{2}$ in. below the bottom of the induction coil and cool the crucible to 1075°C over a period of about 25 min. by operating converter at a very slow rate.

The above cooling procedure helped, but did not entirely eliminate the pipes. Since the remaining pipes were usually near the side of the egg which was nearest the center of the mold a hole was drilled in the center of the mold and connected to each egg mold by sawing through the remaining wall. It was hoped that the center hole would act as a reservoir to supply molten metal to the pipes as they formed and as a heat reservoir to prevent rapid cooling. The final result was that the pipes were reduced somewhat in size and pulled closer to the side of the egg nearest the center of the mold. A top view of the three-hole mold of this type is shown in Figure 16(a).

In the next experiment holes were drilled between the egg molds as shown in Figure 16(b) in order to cut down the cross section of graphite and cause the major part of the heat from the inside of the mold to be transferred down from the center instead of out and then down. The results were good and a large part of the difficulty with pipes was cleared up.

Fig. 16. Cross sections of experimental molds for elimination of pipes.

DECLASSIFIED

240

30

The above work with cooling procedures had only helped the surface condition slightly. The bad surfaces were usually on the side of the egg which was next to the outside of the mold. This could probably be explained by the fact that the minimum thickness of the wall between the outside of the three hole mold, constructed from graphite rod 6 in. in diameter, and the inside surface of each mold cavity was about $3/16$ in. Approximately the same condition existed for four hole molds fabricated from graphite rod 7 in. in diameter. Since about 90 to 95 per cent of the heat from induction is generated in the outside $1/2$ to $5/8$ in. for graphite, there was super heating at this thin wall, and consequently some reaction with the metal. These undesirable surface conditions were nearly eliminated by placing the mold in a $1/2$ in. sleeve. A large part of the inductive heat was generated in the sleeve and more evenly transferred to the mold, thereby eliminating hot spots. This revision is shown in Figure 17.

The difficulties encountered in measuring the temperatures, in loading the crucibles, and in controlling the heating, as well as other experiences gained in casting the metal, prompted a complete redesigning of the casting setup⁽¹³⁾⁽¹⁴⁾⁽¹⁵⁾. The disadvantages previously outlined in using an optical pyrometer were overcome by using a thermocouple to measure the temperatures. In addition, a thermocouple gave a continuous check or record of the heating curve and eliminated the constant attention of one operator using an optical pyrometer. Under casting conditions platinum to platinum-rhodium couples proved unsatisfactory on account of carbonization and alloying from metallic vapors which were liberated at the casting temperatures. After the maximum casting temperature was reduced below 1400°C the inexpensive chromel to alumel couples were tested and proved to be very satisfactory.

DECLASSIFIED

Fig. 17. Graphite egg casting unit with heating sleeve on mold. (1) Viewing chimney, (2) lid, (3) crucible, (4) mold, (5) heating sleeve.

Previously, the crucible and mold had been positioned in the quartz tube and loose insulation packed around them for centering before any uranium biscuits were lowered into the crucible. This arrangement made loading difficult, limited the load by the strength of the quartz tube, and increased the chance of breaking the quartz tube and contaminating the charge with insulation and other foreign materials. For adequate support in the new setup the water-cooled vacuum head was inverted and the charge built up on this head or water-cooled vacuum base which permitted large loads and a marked increase in production. A solid insulation was developed⁽¹⁸⁾ which was placed around the crucible and mold. Then the quartz tube was lowered over this assembly and sealed to the base, thereby reducing the chances of breakage.

The cooling had been controlled by raising and lowering the entire heavy casting assembly. For simplification in the new design, the casting assembly was stationary and a hoist type induction coil was used which could be lowered over the packed assembly and conveniently adjusted to any height for heating.

The redesigned casting unit is shown in Figure 18. A water-cooled base (13) was securely mounted on a welded steel cart (20). The steel cart was mounted on four soft rubber-tired casters (21). The soft rubber tires were important in absorbing small shocks from the uneven floor. Considering the density of uranium and the center of gravity of an uncast charge as shown in Figure 18, the importance of smooth clean floors, a rigid cart supported on soft rubber tires, and careful handling is apparent.

The brass base (13) was cooled by water which passed in through pipe (16) and out through pipe (15). One important feature of this cooling

DECLASSIFIED

Fig. 18. First thermocouple controlled casting unit. (1) Closed-end quartz tube, (2) sillimanite insulation lid, (3) graphite cupola, (4) graphite lid, (5) sillimanite insulation sleeve, (6) induction coil, (7) crucible, (8) thermocouple, (9) thermocouple, (10) mold, (11) thermocouple, (12) insulation base, (13) water-cooled brass base, (14) gasket, (15) water connection, (16) water connection, (17) service tee, (18) vacuum connection, (19) pipe plug drilled for thermocouple, (20) steel cart, (21) rubber tired casters.

DECLASSIFIED

arrangement was that the exit pipe (15) extended nearly to the top of the base and prevented air from collecting in the upper part of the water chamber. A service tee (17) was screwed into the lower side of the base (13). In this work no serious difficulty was encountered with vacuum leaks where two good threads were tightly screwed together with plumber's seal. A short segment of pipe (18) was screwed in the side opening of the service tee to which a 5/8 in I.D. vacuum hose (not shown) was connected. A plug (19) was screwed into the bottom opening of the service tee (17). The plug was drilled for three thermocouple insulators. The above parts were permanently assembled and never taken apart except for repair.

In assembling the unit shown in Figure 18 for casting, the brass base (13) was cleaned and the K-30 insulation brick (12) was placed in position. Then the thermocouple (8)(9)(11) were introduced through plug (19) and adjusted to the correct lengths. These thermocouples were insulated with double-hole aluminum insulators. The thermocouples and their insulators were sealed vacuum tight into the plug (19) by means of Pyseal wax which was carefully flamed over the surface. The mold (10) was next placed in position with care being taken to get thermocouples (9)(11) into their correct positions. The crucible (7) was placed on the mold (10) and thermocouple (8) put in position. Except for the thermocouple holes the crucible, mold, and lid were identical to the unit shown in Figure 13.

The crucible (7) was then packed with crude uranium biscuits. These biscuits weighed from 20 to 30 lbs. each. They were lowered into the crucible by supporting them one at a time on a small wire (18 ga) held with a large pair of pliers. The wire was pulled out after the biscuit was in place. Used chromel or alumel thermocouple wires proved to be very good

DECLASSIFIED

for this operation on account of their ability to undergo sharp flexing without breaking. If a portion of a wire broke off in the crucible the charge was unpacked and the broken piece removed. Various types of mechanical grips were tested and none were found satisfactory because the working space between the crucible wall and the biscuits was too small. This distance was no more than $1/8$ to $1/16$ in.

After packing, the graphite lid (4) and cupola (3) were placed in the position shown. It was found necessary to leave a hole in the lid so that the excess calcium and magnesium could escape quickly or the lid would be lifted and blown out of place. The cupola (3) was placed over the hole in the lid (4) so that the escaping calcium or magnesium vapors would not impinge directly on the sillimanite insulation lid (2) but on the graphite cupola. If the sillimanite insulation lid was directly exposed, these vapors would react with it and cut a hole in it exposing the closed end quartz tube (1). Furthermore, impurities from the sillimanite would fall directly into the charge if the graphite cupola were not used.

The above assembled casting unit was insulated by placing short sillimanite insulation sleeves (5) around it until the top of the last sleeve placed in position projected above the cupola. Then the top was covered with a sillimanite lid (2). The development and the fabrication of the above sillimanite insulation is discussed in detail in a report in Volume 12-B⁽¹⁸⁾. After considerable cracking was encountered with the sillimanite sleeves, the design was changed to a half-sleeve or half-tile which could be built up around a casting assembly in a brick laying manner more quickly than placing sleeves over the top.

DECLASSIFIED

The machined surface on the brass base (13) was carefully cleaned and the rubber gasket (14) placed in position, after which the closed end quartz tube was lowered over the insulated assembly. The rubber gasket (14) was covered with a thin coat of Cello-Seal before assembling. The unit was then moved into position where the pipe (18) was connected to a rubber hose for evacuation. Then the thermocouples were connected to a pyrometer, the water line was connected to the brass base through pipes (15)(16), and a hoist type induction coil was lowered to the correct position for heating.

The mold and crucible were heated above the melting point of uranium and metal dripped or flowed into the mold. In cooling, the coil was raised so that it would heat only the upper one-third of the mold. The induction furnace was operated at such a rate that as large a temperature differential⁽¹⁴⁾ as possible was maintained between thermocouples (9)(11) while the mold was cooling. A temperature gradient of 80°C practically eliminated the pipes; however, the surface condition was not satisfactory. The use of a mold sleeve as shown in Figure 17 had limited the mold to a three-hole mold whereas the elimination of the mold sleeve and the use of 7 in. diameter graphite permitted the fabrication of a four-hole mold and a 33 per cent increase in the number of eggs produced. The urgent need for eggs at this period necessitated the use of the four-hole mold and further work on methods for improving the surfaces.

Although the new design had eliminated pipes and facilitated packing and unpacking, there were still two very undesirable conditions - high carbon content and poor surfaces. The carbon content on the eggs near the surface ran as high as 4000 to 7000 ppm⁽¹⁹⁾⁽²⁰⁾ while the values as low as 300 ppm

DECLASSIFIED

were obtained on the inside of an egg. To correct this condition it appeared that the metal must be run into a cold mold, which would cause a brainy structure under drip casting conditions. To eliminate the brainy structure a valve pouring unit⁽¹⁵⁾⁽¹⁶⁾⁽¹⁷⁾ was designed whereby all the molten uranium would be retained in the crucible until ready for pouring. A cross section of a completely assembled pouring unit is shown in Figure 19. This model had the advantage of retaining all of the metal, in either melting or molten state, in the crucible while the temperature of the mold was well below the melting point of uranium. A carbide lining formed on the crucible and helped to cut down the carbon pickup of the molten metal which was poured into a cold mold where it solidified rapidly and with the formation of little, if any, carbide skin. Analysis of eggs for carbon produced by the pouring method showed that the carbon was not concentrated in the skin as before but evenly distributed throughout the egg. Further, the overall carbon content was much lower on the poured egg. In addition to this improvement, the bad surface condition was automatically improved to the point where practically all eggs were acceptable for their end use.

The use of a valve-pour model, shown in Figure 19, required more accurate graphite fabrication⁽²¹⁾ and the utmost care in assembling. The same cart and brass base are used as shown in Figure 18, except the brass base was altered slightly for thermocouples. Two $\frac{1}{4}$ in. I.D. brass tubes⁽³⁴⁾⁽³⁵⁾ (Figure 19) were installed for thermocouple outlets. The position of the thermocouples in Figure 19 has been rotated into the plane of sectioning and represented as broken lines. Their position in the brass base is not shown in the complete assembly because it would be superimposed on the water inlet and outlet if rotated into the plane of sectioning. A guide bar

DECLASSIFIED

038

Fig. 19. First valve pour egg casting unit. (1) Closed-end quartz tube, (2) sillimanite insulation lid, (3) graphite cupola, (4) graphite lid, (5) graphite crucible extension, (6) graphite crucible, (7) sillimanite insulation half tile, (8) graphite valve button, (9) mold evacuation hole, (10) spillway, (11) graphite push rod, (12) graphite mold, (13) alumundum push rod, (14) thermocouple, (15) thermocouple, (16) mold base, (17) steel socket for alumundum valve rod, (18) guide bar, (19) water-cooled brass base, (20) rubber gasket, (21) water connection, (22) water connection, (23) service tee, (24) vacuum connection, (25) plug, (26) sleeve, (27) steel cart, (28) rubber sleeve, (29) steel push rod, (30) rubber tired casters, (31) evacuation channel, (32) uranium turnings, (33) uranium dioxide, (34) thermocouple outlet, (35) thermocouple outlet.

DECLASSIFIED

240 109

(18) was pressed into the top of the brass base (19). The plug (25) was drilled and a sleeve (26) inserted. Both the sleeve (26) and the guide bar (18) had 1/4 in. holes drilled into them on the center line of the entire unit. This arrangement permitted the 1/4 in. steel push rod (29) to slide easily through these holes. The push rod exit was made vacuum tight by slipping a short segment of rubber tubing (28) over the sleeve (26) and allowing the lower part to extend down over the 1/4 in. push rod (29). When this rubber seal was greased with Cello-Seal, the push rod could be moved up and down very easily. The I.D. of the rubber tube should be equal to or slightly smaller than the O.D. of the push rod. The sleeve (26) was long enough and machined so that a good rigid working fit was obtained for the push rod (29). This type of a push and pull mechanism on a vacuum system has given very satisfactory results in casting and in many other experiments at the Iowa State College Laboratory.

In packing the cart, the thermocouples (14)(15) were set at the correct height and sealed vacuum tight with Pyseal at the point where they projected through tubes (34)(35). One was set at 5/8 in. above the bottom of the crucible and the other at 5/8 in. above the bottom of the mold. The mold base (16) had holes drilled in the correct locations for the push rod and thermocouples. The mold base (16) was designed in two ways in order to reduce the resistance to the flow of gases out of the system. In one case it consisted of two parts with a channel (31) between them and in the other 1/2 in. holes were drilled radially around it. After the mold base (16) was placed in position an alundum rod (13) was placed in the cup (17) on the end of the steel push rod (29). Then the mold (12) was placed in position. The mold base (16) and mold (12) had to be standardized in dimensions

09

DECLASSIFIED

so that the thermocouples would always be at the same height in the mold and crucible. A 1/4 in. graphite rod (11) was fitted in the top part of the push rod hole by slightly sanding with fine emery cloth. It was important that this graphite rod fit closely and move freely. If it was loose, molten metal would flow into the empty space and freeze the graphite rod into the mold, ruining it for further use as well as losing metal.

After the graphite push rod (11) was fitted into the mold, it was cut off so that its top end was 1/8 in. below the graphite valve disc (8) when the crucible (6) and valve (8) were placed in position. The crucible was recessed to a depth sufficient to protect the valve. The valve parts were carefully machined and then ground together on a drill press using slot (36) for rotating the valve. Pouring was accomplished by pushing up the steel valve rod (29) when the uranium was molten. The released valve disc floated to the top of the molten charge.

The crucible had six 3/8 in. holes (9) drilled at an angle in its base to allow the air in the mold to quickly escape upon evacuation. Otherwise, this air would lift the valve and permit small particles to lodge between the ground faces, causing a valve failure. To insure against faulty valves a small amount of uranium dioxide (33) was sprinkled around the edge of the valve to help seal it. A small bunch of uranium turnings (32) was placed on top of the valve⁽¹⁵⁾. These acted as a spring between the valve and uranium biscuits to hold the valve down when the unit was initially evacuated. With a carefully fabricated valve and the absence of small particles to fall on the valve seat, the oxide seal and turnings were not necessary; however, practice showed that their use helped in reducing valve failures from 20 per cent in the early stages of development and 5 per cent in the later stages

DECLASSIFIED

to practically zero. In using uranium dioxide it was important that only small amounts be used otherwise the oxide would be carried through the valve, on pouring, in amounts sufficient to form small oxide pockets in the casting.

The remaining parts were assembled as in Figure 18. In this case larger charges of uranium biscuits were used. Since the charges often protruded above the tops of the crucibles, graphite extension rings (5) were added to enclose the metal. These extension rings, cut from the tops of broken crucibles, represented no cost in graphite. The heating was carried out with an induction coil $12\frac{1}{2}$ in. long. It was set with the bottom of the coil one inch below the bottom of the crucible. This setting would give a "hot top" on the mold. Power was supplied by a 40 Kw Ajax mercury arc converter. A satisfactory casting procedure was as follows:

- (1) Heat at 35 KW until thermocouple (14) (Figure 19) registers 1100°C .
- (2) At this temperature, lower the rate of heating to about 30 KW and heat until thermocouple (14) registers 1170°C .
- (3) Turn off power and allow metal to cool to 1200°C (thermocouple (14)).
- (4) Pour the metal by pushing up on valve rod and turn on the furnace at 10 to 15 KW for 10 min.

The power was cut down in Step (2) to prevent overheating during the fusion period. If the heating was too rapid, the ebullition of impurities was vigorous and some metal was lost over the side of the crucible. Step (4) gave the casting a "hot top" and produced practically pipe-free solid uranium cylinders.

When the mold base (16) was made from K-30 insulation brick the induction coil had to be lowered and raised in steps as heat was supplied to the mold in order to prevent pipes. To promote cooling from the bottom, the mold base (16) was made from graphite⁽¹⁵⁾. Since the mold base was in contact

with the water-cooled brass base and graphite is a fair conductor, the bottom of the mold remained at a low temperature and cooled more rapidly after the uranium was poured. With this design it was possible to obtain pipe-free castings with the procedure given above, i.e. the coil did not have to be moved and the cooling period was much shorter.

Further developments, expansions, and modifications of the above process that have occurred in the production of uranium at the Iowa State College Laboratory will be discussed in another report⁽²²⁾.

Spedding, F. H.

- (1) Spedding, F. H., Wilhelm, H. A., and Gray, C. F. reported in CC-164 (June 20, 1942).
- (2) Spedding, F. H., Wilhelm, H. A., and Gray, C. F. reported in CC-170 (June 25, 1942).
- (3) Spedding, F. H., Wilhelm, H. A., and Gray, C. F. reported in CC-176 (July 2, 1942).
- (4) Spedding, F. H., Wilhelm, H. A., and Keller, W. H. reported in CT-2711 (July 13, 1945) and "Production of Uranium Metal by Reduction of UF_4 by Calcium" reported in E.P.R. Volume 12-B.
- (5) Spedding, F. H., Wilhelm, H. A., and Keller, W. H. reported in CT-2712 (June 26, 1945) and "Production of Uranium Metal by Reduction of UF_4 by Magnesium" reported in E.P.R. Volume 12-B.
- (6) Spedding, F. H., Wilhelm, H. A., and Gray, C. F. reported in CC-177 (July 9, 1942).
- (7) Spedding, F. H., Wilhelm, H. A., and Gray, C. F. reported in CC-205 (July 16, 1942).
- (8) Spedding, F. H., Wilhelm, H. A., and Gray, C. F. reported in CC-238 (August 15, 1942).
- (9) Spedding, F. H., Wilhelm, H. A., and Gray, C. F. reported in CC-258 (September 15, 1942).
- (10) Spedding, F. H., Wilhelm, H. A., and Gray, C. F. reported in CC-298 (October 15, 1942).
- (11) Spedding, F. H., Johns, I. B., Warf, I. C., and Newton, A. S. reported in CC-580 (April 15, 1943).
- (12) Spedding, F. H., Wilhelm, H. A., and Butler, T. A. reported in CT-456 (No date).
- (13) Spedding, F. H., Wilhelm, H. A., and Gray, C. F. reported in CT-393 (No date).
- (14) Spedding, F. H., Wilhelm, H. A., and Gray, C. F. reported in CT-422 (January 15, 1943).
- (15) Spedding, F. H., Wilhelm, H. A., and Gray, C. F. reported in CT-490 (February 15, 1943).
- (16) Spedding, F. H., Wilhelm, H. A., and Gray, C. F. reported in CT-542 (March 15, 1943).

DECLASSIFIED

~~CONFIDENTIAL~~

- (17) Spedding, F. H., Wilhelm, H. A., and Gray, C. F. reported in CT-609 (April 20, 1943)
- (18) Spedding, F. H., Wilhem, H. A., Calderwood, W. and Gray, C. F. "Radiation Shields and Their Fabrication" reported in M.P.R. Volume 12-B.
- (19) Spedding, F. H., Warf, J. C., Tevebaugh, A. D., and Fassel, V. A. reported in CC-432 (January 22, 1943).
- (20) Spedding, F. H., Warf, J. C., Tevebaugh, A. D., and Fassel, V. A. reported in CC-470 (January 22, 1943).
- (21) Spedding, F. H., Wilhelm, H. A., Hull, E. and Gray, C. F. "Machining of Graphite and Special Operations" reported in M.P.R. Volume 12-B.
- (22) Spedding, F. H., Wilhelm, H. A., Thomson, R. E. and Gray, C. F. "Present Practices in the Uranium Foundry at Iowa State College" Reported in M.P.R. Volume 12-B.

240-045

~~CONFIDENTIAL~~

DECLASSIFIED