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RADIATION SHIELDS AND THEIR FABRICATION

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RADIATION SHIELDS AND THEIR FABRICATION*

Work done by F. H. Spedding, H. A. Wilhelm, and W. Calderwood

Paper written by: W. Calderwood and C. F. Gray

The vacuum casting of larger amounts of crude uranium at the Ames Laboratory and the necessity of controlling the casting operation to a greater degree brought about the change from a crucible-mold assembly supported on loose insulation in a closed end quartz tube covered with a water-cooled vacuum head, to a casting unit where all parts are supported on a water-cooled vacuum base and covered with a closed end quartz tube. This change made some type of solid insulation necessary. A desirable solid insulation for use in the above new casting unit should have the following properties.

- (1) Solidity and sufficient strength that a piece $\frac{1}{2}$ inch thick will stand ordinary handling without breaking.
- (2) Freedom from crumbling and dust.
- (3) Ability to withstand temperatures of 1500°C in contact with graphite.
- (4) Sufficient insulation and radiation shielding ability to protect the quartz tube and hold the major part of the heat generated by inductive heating.
- (5) Ability to stand high temperature gradients and be resistant to thermal shock.
- (6) Resistance to warping under the strain of successive heating and cooling.

An investigation was started at Iowa State College Laboratory to develop an insulation possessing the above properties for the thermocouple-controlled, valve-pouring casting unit, which is fully discussed in other reports.^{1,2}

*Based on CT-393, CT-609, CT-686, CT-816.

The problem was first attacked by attempting to coat the inside of a closed end quartz tube with a layer of insulation refractory. Silica was selected as a possible liner, because of its use in general foundry practice. Sea sand and boiled linseed oil were mixed according to common foundry practice into a thick paste and applied to the inside of a quartz tube. It was necessary to paint the quartz with linseed oil before the sand mixture would stick. The sand adheres firmly to the quartz tube after it was baked for one hour in an electric muffle at 200°C, but when the baking temperature was raised to 900°C the binder was completely destroyed and the sand liner crumbled.

Further work with liners was then discontinued and the possibility of fabricating a refractory sleeve, about $\frac{1}{2}$ in. in thickness and of the correct dimensions to fit between the graphite crucible-mold and the quartz tube, were investigated. Thermal-setting sillimanite cement, obtained from General Refractories Company of Danville, Illinois, was selected as a possible refractory. This cement is a dry powder which varies in particle size between 100 and 300 mesh. Its color is reddish brown, indicating an iron impurity. The work "silimanite" refers to one of the alumina minerals and is often confused with andalusite and kyanite. Kyanite is the most plentiful of the three. Adequate supplies of true sillimanite for industrial use were not at this time known to be available in this country. All three minerals decompose on heating to mullite ($3\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$) and glass. The decomposition temperature ranges are from 1100°C to 1410°C for kyanite, 1410 to 1500°C for andalusite, and 1550 to 1625°C for sillimanite. Whatever the true composition of the sillimanite cement obtained from General Refractories, tests showed that a thick paste made with water can be formed in various shapes and baked at 1000°C to give a hard, noncrumbling refractory that will stand temperatures up to 1500°C in contact with graphite.

In order to reduce the rate of conduction of heat through the insulation, an attempt was made to produce a porous refractory³ by mixing a carbonaceous material with the sillimanite cement, which would burn out on firing. The first carbonaceous material tested was Shawinigan black (acetylene black). It was mixed with sillimanite and water to form a homogeneous slip. An insulation and radiation sleeve was formed by casting this slip in a plaster-of-paris mold. Large cracks developed in the walls of the sleeve while it dried in the mold and the Shawinigan black did not burn out readily from pieces of the sleeve when heated over a Meeker burner for long periods.

In the next experiment Shawinigan black was replaced with Norblack, which was a coarser form of carbon. It is possible that Shawinigan Black is so fine that it coats the sillimanite particles to such an extent that they lose their cohesive power. A slip was prepared with Norblack in the same manner as before and cast into the same plaster-of-paris mold. This sleeve did not crack upon drying but did break on removal from the mold. In another experiment using the same slip, the mold was dusted with talc ($3\text{MgO} \cdot 4\text{SiO}_2 \cdot \text{H}_2\text{O}$) before the slip was cast, and in this case the sleeve was removed without cracking or breaking. Further tests favored the use of talc dusting and as a result it became standard practice. The sleeves, removed in one piece, were dried and then fired at 650° to 700°C . The fired sleeves were fairly porous but had very low strength.

Another technique of forming sillimanite sleeves was tested. A round glass form of the correct size was dipped into a sillimanite Norblack slip, allowed to dry, redipped and again allowed to dry until a layer of sufficient thickness had been built up on the form. Difficulty was experienced in removing the sleeve from the form after drying. One method developed for removing it was to cover the glass form with a light oil and a light weight piece of cloth. After the sleeve had dried the glass form could be slipped out and the cloth unwrapped from the inside. Sleeves prepared in this manner were fired at 650° to 700°C . The shrinkage due to firing cracked all of the sleeves into several pieces. This method was discontinued.

In all of the previous experiments one of the more serious difficulties was the cracking of the fired sleeves due to shrinkage. It was suggested that if the porosity could be radically increased, the shrinkage from firing might be absorbed by the porous structure without breakage. To increase the porosity, a large amount of wood sawdust was substituted for the Norblack in the slip. Such a sawdust-sillimanite slip was poured into a plaster-of-paris mold. Difficulty was experienced with the excess water draining away or being absorbed by the mold. However, with great care, the sleeve could be removed in one piece, dried, and fired at 700°C in an electric muffle. The fired piece was extremely brittle and highly porous. Another important observation at this point was that the fired piece shrank only very little, indicating that high porosity reduces warping and shrinkage.

In order to reduce the porosity and increase the strength, the sawdust-sillimanite ratio was set at 1 to 1 by volume. Only enough

water was added to give a semi-plastic mass. The sleeve was formed by tamping the semi-plastic mass into the annular space between two concentric steel tubes, but after formation it was impossible to remove the sleeve. Another test with only a moist 1 to 1 mixture tamped into the mold gave the same results. This difficulty with the mold prompted the change in mold design to a plaster-of-paris outside and a light, tinned, sheet steel inside. The plaster-of-paris outside was cut into halves and could be removed from a tamped sleeve without difficulty. The tinned core could be removed, without breaking the sleeve, only with difficulty. Sleeves formed from tamped moist 1 to 1 mixture in this mold would stand by themselves. They were dried at 110°C and then fired. The fired cylinders were porous, strong enough for careful handling, and low in resistance to abrasion. Another sleeve produced in a similar manner from 1 to 2 sawdust-sillimanite mixture was noticeably improved in strength and resistance to abrasion.

The partial success in the fabrication of an insulation-radiation sleeve brought about the production of enough sleeves for a trail run (without uranium metal) on the regular production equipment. These sleeves were 6.5 in. I. D., 7.5 in. O. D., and 5 in. long, with the wall thickness varying between 0.375 and 0.50 in. They were packed around the 6.25 in. graphite production crucible-mold unit, covered with a quartz tube, evacuated, and heated until a thermocouple in the graphite crucible indicated a temperature of 1400°C. The insulation properties were good in that the rate of heating was rapid and no part of the quartz tube became visibly hot during the entire run, except that the position of the junctions of the sleeves could be detected through the quartz tube above 1300°C. Cooling and unpacking revealed that every sleeve had about two cracks parallel to its axis and approximately opposite to each other. Otherwise the results appeared to be very satisfactory. The positions of these cracks indicate that some strong stresses were present in the sleeve during the heating and that these stresses might be relieved by cutting the sleeves into two pieces parallel to their axis, with the cuts opposite to each other. This idea was tested and proven correct in runs in which uranium was cast.^{3,4,5} The parallel cuts are made oblique to the radius of the sleeve in order to prevent the radiation of the crucible from striking the quartz tube. Further experiments showed that small cracks can be repaired with air-setting sillimanite cement.

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From the above results a new mold for forming half sleeves was designed. It is shown in Figure 1. The mold is made in two sections, one of which forms the outside of the half-sleeve and the other the inside. Both parts are made from plaster-of-paris. Figure 1(A) shows the outside mold, and Figure 1(B) the inside mold. The inside mold has a wooden frame into which the outside mold fits to insure a uniform space between them for tamping the moist sawdust-sillimanite mixture. The two parts are rigidly clamped together before the mixture is tamped into the mold. Two views of the assembled mold are shown in Figures 1(C). To provide enough sleeve for trimming obliquely to the radius, the molds are designed to form a sleeve that projects tangentially 0.50 in. beyond one half of a circle. The half-sleeves are trimmed so that two of them fit together to form a circular sleeve as shown in Figure 2.

In the actual production of half-sleeves or radiation shields, the mold is dusted with talc and assembled, and a moist 1 to 2 sawdust-sillimanite mixture is carefully and evenly tamped into place. Uneven tamping produces straitions and weak boundries in the final product. When the mold is filled, the interior section is removed first. The half-sleeve is then loosened from the outside mold with a thin spatula, pushed free from the exterior section, and stood on a board. On account of its low strength the half-shield is kept on its board until completely dried in a 110°C oven. After drying, the half-shields are handled manually. They are then stacked in an oilfired car kiln and fired at 1100°C, the temperature which proved most satisfactory as determined by a series of tests. After cooling, the edges and ends of each half shield are trimmed with a hack saw blade in a specially built miter box so that any two half-shields will fit together to form a sleeve 7.5 in. O. D., 6.5 in. I. D., and 5.0 in. long. The ends are trimmed perpendicular to the axis of the sleeve. The edges are cut obliquely to the radius to prevent radiations of the crucible from striking the quartz tube as mentioned above.

The process described above for the fabrication of radiation half-shields or half-sleeves is satisfactory for limited production; however, the increased production rate⁶ for casting uranium makes the method obsolete for the following reasons:

(1) The thickness of the walls of the shields increases slightly each time the mold is used, from the abrasion on the plaster-of-paris by tamping the sawdust-sillimanite mixture into the mold with a wooden

stick. Since the space between the quartz tube and the graphite crucible is limited, a small increase in thickness can render the shields useless.

(2) Too many man-hours are required per shield.

(3) Although talc is used on the mold each time, it becomes continually more difficult to separate the half shield from it. Many shields are lost in removal from the mold.

(4) Since each shield has to be set on a separate board for drying, too much space is used for this operation.

(5) The fired sillimanite is very abrasive and trimming with hack saw blades is inefficient and wasteful of blades.

A thorough study of these problems brought about many improvements in the process. The mold was redesigned so as to eliminate the tamping procedure. The newer one which is shown in Figure 3 consists of a plaster-of-paris mold, two wooden guides, and a clamp to hold the guides and mold in a fixed position. The plaster-of-paris mold shown in Figure 3 (A) forms the outside of the half-shield. The circular surface on the wooden guide shown in Figure 3 (B) has the same radius as the inside of the half-shields. When the guides and the mold are clamped together as shown in Figure 3 (C) a half-shield is formed by packing the moist sawdust-sillimanite mixture on the inside surface of the mold and the two guides are used for leveling the inside surface of the half-shield. The complete procedure for forming a half-shield is as follows:

(1) Clamp guides and mold together so that axes coincide and the top surfaces lie in the same place.

(2) Dust mold with talc.

(3) Mix sawdust and sillimanite cement in the ratio of 1 to 4.

(Later tests proved that a ratio of 1 to 4 produces better shields than the 1 to 2 ratio).

(4) Roughly place the sawdust-sillimanite mortar in the mold with a trowel.

(5) Press this mortar into place and level the inside surface of the half-shield by passing a rolling pin over the inside surface, using the end guides to control the thickness of the shield.

(6) Add mortar where any deficiency exists and remove any excess with trowel.

(7) Remove guides and set mold aside for partial drying. One hour or longer is required, depending upon the humidity.

(8) Before the half-shield is completely dry, but as soon as it is dry enough to stand of its own strength, loosen it from mold with a spatula and slide it onto a tray. Shields tend to crack if drying proceeds too far in the mold.

(9) Place tray of partially dry half-shields in oven and completely dry at 90°C.

(10) Stack dry half-shields in kiln and fire to 1100°C as rapidly as kiln can be heated.

(11) After cooling to room temperature, place in special jigs on a Clipper Masonry Saw and trim to required dimensions.

For pressing and leveling the mortar, rolling pins of glass, steel, wood, and felt-covered wood have been used. The felt-covered wood rolling pin gave the best results. Wetting the felt before rolling definitely improves the results and prevents the sawdust-sillimanite mixture from sticking to the rolling pin. This revised mold together with the forming operation greatly increased the rate of production. The use of the Clipper Masonry Saw for trimming the half-sleeves increases production a great deal as well as giving smooth and true cuts. In fact, the edges of the half-shields are now trimmed perpendicular to the axis instead of obliquely because they fit together well enough that only a negligible amount of radiation strikes the quartz tube.

When the large 15 in. induction furnace was put into use for casting uranium larger insulation was required. Practice showed that it was necessary to form the larger sleeves into three parts, or "third-sleeves." The larger sleeves are fabricated in exactly the same manner as the smaller half-sleeves described above.

After an examination of the porosity in the half-sleeves it was noted that breakage usually occurs at points having very large voids which were caused by large pieces of sawdust. To eliminate this, all sawdust was sized through a screen having $\frac{1}{4}$ in. openings and that retained on the screen discarded. Half-sleeves fabricated with sawdust which passes a $\frac{1}{4}$ in. were stronger after firing, but the working properties of the mortar was not as good. Further work showed that the very fine sawdust also interfered with the working properties of the mortar. Using only that portion of sawdust which passes a $\frac{1}{4}$ in. screen but was retained on a 40 mesh screen gave the mortar good working properties and the fired product good strength.

In order to increase the porosity and retain the fired strength, a study was made on the possibility of increasing the percentage of sawdust and adding a plastic or flux bonding agent such as A. P. Green dry-milled fire clay or Nepheline Syenite ($\text{Na}_2\text{O} \cdot \text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$). With the latter, a strong porous structure was obtained when a sawdust-to-sillimanite ratio of 2 to 3 was fired at 1100°C; however, such half-sleeves soften

during use. The addition of 10 per cent fire clay increased the plasticity. The best results for fire clay was obtained with a sawdust-sillimanite ratio of 1 to 3.

A study of the maximum firing temperatures showed that strength increases with the maximum firing temperature. At 1000°C, the half-shields were brittle and weak. At 1100°C they were much stronger and could be cut with a Clipper Masonry Saw without difficulty. At higher temperatures, the strength and hardness increased and as a result they could be cut only with great difficulty on the masonry saw. Sillimanite transforms to mullite ($3\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$) and Cristobalite (SiO_2) at 1450° to 1550°C. Mullite is a strong refractory and softens above 1600°C.

For some special work, some radiation half-shields were needed that could stand much higher temperatures. For this, Norton's Alundum Cement No. 345 was selected as a possibility. The alundum was mixed with sawdust and water to give a thick mortar, and worked into a half-sleeve mold with a trowel and rolling pin. It was much more difficult to handle than sillimanite. The alundum half-shields could not be removed in one piece from old molds and only with difficulty from new molds, well powdered with talc. They dried very slowly, cracked easily, and were quite brittle when handled. However, further work developed a procedure by which alundum half-shields could be produced and fired. The mold was lined with paper and filled with slightly dampened alundum cement. The sawdust was eliminated. The damp alundum was rolled in to place under great pressure, and after rolling, more water was added with a paint brush. This final addition of water helped to bind the damp particles together, and, when added at this point, prevented the alundum from sticking to the roller. These half-shields were strong enough after drying for transferring to the kiln. They were fired at 1100°C to give a fairly strong half-shield that could be easily trimmed on a masonry saw. When tested, the alundum half-shields gave good results up to 1800°C; however, sustained heating above 1900°C caused softening, warpage, and cracking. Since pure Al_2O_3 melts at 2050°C; the softening at 1900°C was probably due to the binders in the cement.

In summary, a satisfactory sillimanite insulation-radiation shield was developed that could be easily fabricated and easily assembled and disassembled. For other high temperature experiments, an alundum insulation-radiation shield was developed which was satisfactory for continuous use up to 1800°C.

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(A) OUTSIDE MOLD

(B) INSIDE MOLD

(C) ASSEMBLED MOLD

Fig. 1. Mold for forming insulation half sleeve: (A) outside mold, (B) inside mold, (C) assembled mold.

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Fig. 2. Method of trimming and fitting half sleeves together.

Fig. 3. Improved mold for forming half sleeves: (A) plaster-of-paris mold, (B) wooden guide, (C) assembled mold.