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Lawrence Livermore Laboratory

ENCAPSULATION OF UC-PuC MIXTURE FOR CALORIMETRIC STUDY

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December 28, 1976

MASTER

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Prepared for U.S. Energy Research & Development Administration under contract No. W-7405-Eng-48.

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ENCAPSULATION OF UC-PuC MIXTURE FOR CALORIMETRIC STUDY

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ABSTRACT

Tantalum capsules lined with tantalum carbide were loaded with a mixture of uranium carbide-20 wt% plutonium carbide and sealed with an electron beam weld for use in Argonne National Laboratory calorimetric studies of reactor fuels. Experimental welding tests were made to determine optimum welding parameters. Special equipment available only at LLL was used and techniques were developed to perform the loading and welding operations without exposure to moisture or oxygen and with complete containment of the radioactive particles. Three capsules were successfully loaded, welded, tested, and delivered to ANL.

INTRODUCTION

In 1975, we were requested by Argonne National Laboratory to load tungsten capsules with a mixture of uranium carbide-20 wt% plutonium carbide in a super dry oxygen-free (less than 5 ppm oxygen) atmosphere and seal these capsules by welding and maintain a high vacuum inside the capsules. These capsules were to be used in a calorimetric experiment on prospective reactor fuels at ANL; and it was essential that the capsules contain a high vacuum to prevent excessive pressures in the capsules during heating. Six capsules were loaded, welded, and delivered to ANL as a result of this request. Calorimetric studies of the material at temperatures below the melting point were made successfully, but when the carbides were heated into the molten range, a reaction apparently took place between the molten carbides and the tungsten capsule and leaks resulted. Subsequent investigation at the Los Alamos Scientific Laboratory indicated that tantalum carbide would resist the molten carbides. On this basis, work was started to prepare tantalum carbide-lined tantalum capsules for further calorimetric work.

We were requested by Donald F. Fischer of ANL to load and seal these capsules, and we again agreed to the request because LLL is the only

facility in the ERDA complex with both a plutonium contaminated glove box in which the required purity of atmosphere could be obtained and maintained and which has a plutonium contaminated electron beam welder in which to do the welding. The work was accepted on a time available, no priority basis.

MANUFACTURE OF CAPSULES

The capsules were prepared by Ultramet of Paicoma, CA, by chemical vapor depositing (CVD) tantalum carbide on a mandrel to about 0.1 mm-thick, chemical vapor depositing pure tantalum on top of the tantalum carbide to a thickness in excess of 1 mm, and then machining the surface to give a 1 mm-thick tantalum wall. The mandrel is then removed. A drawing of the finished capsule is shown in Fig. 1. Note the hollow center post in the capsule. This feature is to permit placing a thermocouple in the center of the mixture during the calorimetric tests, and its location caused some difficulty both in making the capsules and in filling the capsules with the UC-PuC mixture. The cap is also coated with TaC and is electron beam welded in place.

When this type of capsule was proposed for welding, we questioned the detrimental effect of the tantalum carbide in the weld joint because our experience in welding dictated that a ceramic coating would disrupt the welding action. Indications from the supplier were that the joint area was to be free of tantalum carbide, and its influence on welding would be insignificant. Since the cost and difficulty of obtaining the lined capsules precluded using the tantalum carbide coated capsules to determine optimum welding parameters, we agreed to do our procedure development on simulated capsules made from pure tantalum tubing of a smaller diameter and on end caps made from bar stock.

UC-PuC PELLETS

The mixture of uranium carbide/20 wt% plutonium carbide pellets were prepared at the Los Alamos Scientific Laboratory. The pellets were compressed to a dimension of approximately 12 mm diameter and 6 mm thickness. They were then loaded into a stainless steel tube, the end welded shut with a protective atmosphere inside, and shipped to ANL. They were delivered to us in the welded tube as prepared at LASL. The pellets could not be made

in doughnut shapes with a hole to accommodate the center post, so the pellets had to be broken into pieces small enough to fit between the post and side wall of the capsule.

WELDING PROCEDURE DEVELOPMENT

The ANL requirements for the weld were to achieve at least 90% penetration, eliminate cracks or other defects, withstand the heating and cooling in the calorimetric apparatus, and not disturb the UC-PuC mixture. These requirements had been met on an earlier order for tungsten capsules.

Six short tubes of tantalum, machined to simulate the capsules, and six caps machined from bar stock were received from ANL for parameter development. These tubes were smaller in diameter (12.7 mm) than the final capsules (14.3 mm), and we found that six were not enough for both development and verification samples. These tubes were used to determine parameters, and a sample was sent to ANL for approval. This sample was welded at: 0.762 m/second travel speed, 70 kV accelerating voltage, 7.5 mA beam current with the beam defocused to give a 0.5 mm spot at the work. The filament current was first reduced to 1.75-1.8 A to give a soft, low-powered spot, and this was applied to the joint for 2 revolutions to preheat the part. The filament current was then brought up to 2.1 A, and three welding passes made at full power.

A copper chill fixture was machined to fit tightly around the 6 mm tubing and positioned 1.5 mm below the weld joint. The purpose of the chill fixture was to keep the welding heat from affecting the character of the mixture. The absorption of heat by the chill fixture must be compensated for by adding heat during welding to obtain the desired penetration, and, therefore, the fixture had to be used in development welding even though the tubing was empty.

After the initial work was done and the sample sent to ANL, some tantalum capsules and lids with the same approximate dimensions as the practice capsules were found surplus at LLL, and these were used to make verification welds. After making several repetitive samples, we found that the penetration was insufficient at the stated set of parameters, and that it was necessary to raise the current to 10 mA to obtain the desired penetration.

The tantalum carbide-lined capsules were received from ANL, ultrasonically cleaned in MEK (methyl ethyl ketone) as specified, and mass spectrometer helium leak-tested. All were tight except for capsule #1, which had a large leak. Capsule #2 had been specified by ANL as the one to be used for weldability checking and as a sample capsule to use for calibrating the ANL calorimetric apparatus. This empty capsule, containing a thin tungsten circular wafer and used as a heat shield between the material and the cap weld, was placed in the chill fixture and into the non-contaminated welder to check procedures and parameters. The cap was tack welded in 4 spots to hold it in place after the restraining bar was removed. Some weld pool eruptions were observed during the tack welding. When the finish weld was made, one side of the joint welded smoothly, but eruptions occurred on the other side of the joint and molten metal was expelled. This left the surface of half the weld with a rough and lumpy appearance. This was thought to be caused by the TaC on the capsule surface because almost all the TaC had been ground away from the cap edge in order to fit it into the capsule. Welding was done at the parameters stated previously, except that the amperage was raised from 7.5 mA to 10.0 mA. No measure could be taken of the penetration but the capsule was helium mass spectrometer leak-checked and found to be leak tight. It was sent to ANL for evaluation with our comments on the welding action.

It was decided by ANL that capsule #1, the leaking capsule, should be welded in the contaminated machine to check the transfer of parameters to this machine and a further test of the weldability of the TaC lined material. When this capsule was welded, large expulsions of molten metal occurred and several holes were left in the weld. The capsule did not appear to be of the best quality, and the poor welding action seemed to be caused by a combination of the TaC in the joint and poor material. There were no spare parts, so the decision was made to fill and weld the remaining capsules as best we could. Since the TaC had been removed from the cap of capsule #2, we removed the TaC from the edges of the remaining caps by rotating the cap in a jeweler's lathe and holding an abrasive stone against the edge of the cap. This removed most of the TaC.

LOADING

The loading of the capsules was performed by Mr. W. Thayer in a special plutonium-contaminated glove box in which a very dry and pure atmosphere could be obtained and maintained. This atmosphere was required because of the susceptibility of the mixed carbides to oxygen and water vapor. The oxygen content of the glove box was kept below 2 ppm at all times and the moisture, hydrogen, and nitrogen level was kept very low by suitable trains. The tools, pellet tube, and capsule #3 were loaded into the vacuum lock of the box and the lock evacuated for one hour before transferring the contents to the glove box. This evacuation removed atmospheric contamination from these items and maintained the glove box atmosphere below 2 ppm oxygen.

The capsule was removed from its container, and with its cap and a thin tungsten heat shield was weighed to within 0.0001 g on the Mettler H-30 in the glove box. The stainless steel tube containing the pellets was then opened with a sharp tubing cutter and the pellets removed as necessary. Since the pellets were too large to fit inside the capsules with the center post, a Plattner's mortar and pestle was supplied by ANL. A pellet was placed in the mortar and broken with a sharp hammer blow. The large pieces were picked out with tweezers and placed in the capsule, while the fines and powder were poured into the capsule using a foil funnel. A camel hair brush was used to ensure that all particles of material were removed from the mortar and pestle and brushed into the capsule. The weld joint area was carefully brushed to remove any particles of material that might interfere with welding. When the capsule was filled with material, a thin tungsten heat shield was placed on top of the material to protect it from direct radiation heating during welding, the cap set in place, and the assembly again weighed to 0.0001 g. The difference between weights was the weight of material; approximately 40 g.

The loaded and weighed capsule was placed in the copper chill fixture and a retaining bar fastened across the end of the fixture. This bar held the two halves of the fixture together and also held the end cap firmly in place. The capsule and chill fixture was placed in a plastic container, and the top sealed with tape. The contaminated container with

the capsule was then bagged out of the super dry glove box by Hazards Control and into the electron beam welder glove box. It was a requirement that the capsule be kept in an upright position at all times after the heat shield was in place to prevent any material from getting above the heat shield. Care was taken during all the bagging and moving operations so that at no time was the capsule turned from the vertical position.

The glove box enclosure around the welder had been purged with dry nitrogen for an hour before the plastic container with the capsule was bagged in. The container was opened, the chill fixture with the capsule placed in the rotating welding fixture, and the assembly moved into the welding chamber. The chamber was evacuated; the cap was tack welded in four places to hold it securely, the chamber opened and the retaining bar removed from the chill fixture, and the assembly returned to the welding chamber. The welding chamber was then evacuated overnight to ensure a high vacuum in the capsule, and the final weld was made the next morning. Vacuum in the welding chamber before welding was at 3×10^{-5} torr, or better.

The vacuum in the welding chamber was then broken with helium gas and the capsule kept in the helium atmosphere for at least 15 minutes. If there is a leak in the capsule weld, the vacuum existing inside the capsule will pull helium into the capsule through the leak and the vacuum will be reduced. When the outside of the capsule is evacuated during leak testing, this helium will be pulled out through the leak and signal the leak detector. This is our standard method of leak testing sealed capsules.

After welding, followed by soaking in helium, the capsule was again bagged into the super dry atmosphere glove box and reweighed. It was also visually examined with a low power microscope for defects in the weld. After all the capsules had been loaded, welded, weighed, and examined, they were then helium mass spectrometer leak-tested. They were all placed in a contaminated container, the container evacuated, and placed on the leak detector. If a helium signal had been received, the capsules would then have been tested individually to determine the leaking capsule. All capsules were leak tight, however, so the contaminated capsules were placed in their containers, taking care not to contaminate the outside of the containers,

double bagged for additional protection, sealed in a tin container, and shipped to ANL in the upright position.

WELDING OF CAPSULES

The first loaded capsule to be welded was capsule #3. Because of the difficulty encountered in welding capsule #1, the welding procedure was changed. The changes were to increase the heat and melt more of the tantalum. The object was to remove the support for the tantalum carbide, causing it to break up into small pieces under the action of the beam and reduce the disruptive effect upon the weld. The increase in heat was obtained by raising the beam focus to a spot 25 mm above the work to give a more diffuse beam, slowing the travel speed to 0.508 m/s from 0.762 m/s to compensate for the reduced penetration resulting from the defocused spot, and make three preheating passes instead of two.

When the cap was tack welded in place, a reaction occurred at one of the tack welds and molten metal was expelled from the joint. When the full power welding pass was made, a similar reaction occurred at the same spot, and more material blew out. It appeared to be caused by tantalum carbide from the lining in the joint, rather than carbide from the mixture. The weld was low at that spot, but it was difficult to see if there was a hole in the weld by looking through the optics of the welder. When the capsule was removed from the welding chamber into the glove box enclosure and it could be examined more closely, we could see that there was a hole in the weld.

The capsule was returned to the welding chamber and an attempt was made to flow the edges of the hole together by melting the edges of the hole with the beam. This attempt was unsuccessful. Tantalum wire was obtained and an attempt made to place the wire in the hole, melt the wire in place, and fill the hole. This technique is used in our normal operations, but the wire or other material must be fastened in the hole, usually by resistance spot welding, to prevent it from coming out of the hole when the beam hits it. This could not be done in the contaminated welding chamber, and the wire kept coming out of the hole when struck by the beam. Several unsuccessful attempts were made to repair the hole; then the electron gun filament burned out, stopping the welding operation. The capsule was removed to the super

dry atmosphere glove box while the filament was being replaced, on eight hour operation. The decision was then made to continue with the remainder of the capsules and hope we could get at least one good one.

Because of the difficulty found in welding capsule #3, an attempt was made to remove the tantalum carbide from the inner surface of capsule #4 at the weld zone. The capsule was mounted in a lathe and a machine-mounted high-speed grinder used. This was not successful, nor was a hand held low-speed grinder, because the carbide coating was too hard. The capsule was recleaned and loaded in the specified manner.

Because of the difficulty with capsule #3, the procedure was changed to use a longer preheat cycle of 4 revolutions, instead of 3. Just before the completion of the weld, an expulsion of metal occurred and a depression could be seen. Although we were not sure that a hole was present, the operator preheated the part again and went back over the hole with a lower power beam 9.5 mA. The depression filled to the extent that we were sure that there was no hole present. The operator inadvertently backfilled the chamber with dry nitrogen, but he did not open the chamber; he then re-pumped the chamber to 2.5×10^{-5} torr and backfilled with helium. On withdrawal from the chamber, the weld looked good with no visible hole. Metal loss during welding was 0.078 g.

Capsule #5 was welded in the same manner, except that six preheat revolutions were used instead of four. The weld looked very good with no metal expulsions and no depressions. The metal loss during welding was 0.046 g.

Capsule #6 was welded in the same manner as #5. There was a minor expulsion at one of the tack welds and several minor expulsions during welding but no depressions or holes. The weld looked good, and the metal loss during welding was 0.048 g.

After completion of capsule #6, an attempt was made to close the hole in capsule #3. Tantalum wire was placed in the hole in both wire form and in ball form, but the hole could not be closed. In the process of attempting the repair, several other holes opened up and the repair attempt was abandoned. The action of the molten metal in the capsule during the repair attempt and during the tack welding of the capsules leads us to

believe that the welding difficulty may be caused by defect areas in the capsules and not by carbide in the joint. When we questioned the manufacturer of the capsules; he admitted that they had not actually welded any tantalum lined capsules, but they had assumed from the characteristics of the material that there would be no trouble.

This leaky and badly contaminated capsule was placed in an available stainless steel container, the container electron beam welded shut, leak tested, and returned to ANL with the other capsules.

SUMMARY

This work shows that tantalum carbide-lined tantalum capsules can be successfully welded, if the proper technique is used. Proof of this is the fact that 3 capsules were welded with no leaks or other defects. The work is not complete, however, since the reason for the reaction of certain areas in the capsules to the beam with subsequent expulsion of metal has not been clearly determined. If further work of this type is to be undertaken, then sufficient tantalum carbide coated capsules or tubes must be provided for welding development purposes to positively determine the cause.

This work also shows that LLL has the capability for safely handling and encapsulating material that is highly radiotoxic and susceptible to contamination by small amounts of oxygen and moisture.

TANTALUM CAPSULE AND CAP

Figure 1

NOTE: MEASUREMENTS INCLUDE
THE 0.004 T&C LAYER
WHERE APPLICABLE.

DRAWING : CFN-C-1
DRAWN By : D. FISCHER
DATE : 5/6/75

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Printed in the United States of America
Available from
National Technical Information Service
U.S. Department of Commerce
5285 Port Royal Road
Springfield, VA 22161
Price: Printed Copy \$; Microfiche \$3.00

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