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REMOTE MICRO-ENCAPSULATION OF
CURIUM-GOLD CERMETS

MASTER

M. S. Coops
A. L. Voegelé
W. N. Hayes
D. H. Sisson

Lawrence Livermore National Laboratory
Livermore, California 94550

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M. S. Coops
A. L. Voegelé
W. N. Hayes
D. H. Sisson

Lawrence Livermore National Laboratory
Livermore, California 94550

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INTRODUCTION

We have developed a technique to fabricate miniature, high-density capsules of curium-244 oxide contained in three concentric jackets of metallic gold (or silver), with the outer surface being free of alpha contamination. The completed capsules are right circular cylinders 0.2500-inch diameter and 0.125-inch tall, with each level of containment soldered (or brazed) closed. A typical capsule would contain ~70 mg of ^{244}Cm (5.7 Ci) mixed with 120 mg of gold powder in the form of a cermet wafer clad in three concentric, 0.010-inch thick, liquid tight jackets. This method of fabrication eliminates voids between the jackets and produces a minimum size, maximum density capsule. Cermet densities of 11.5 g/cc were obtained, with an overall density of 17.3 g/cc for the finished capsule.

The completed capsules are dimensionally stable for periods exceeding five years, and show no signs of radioactive leakage during that period of time. The stability of the package is attributed to the method of fabrication of the gold components. The component capsules are made by electroplating gold on precision-machined pure copper mandrels, machining the plated surface to size, and chemically dissolving the mandrel. This fabrication technique was chosen for simplicity of manufacture and also has the advantage of creating a microporous gold structure that allows the helium gas produced by alpha decay to diffuse through the containment structure while retaining all particulate materials.

DESCRIPTION OF CONTAINMENT ASSEMBLY

Our choice of package dimensions was dictated by experimental needs and could be varied to suit other configurations. Our outer (final) capsule is a right circular cylinder held to precisely 0.2500-inch diameter

by $0.125 \pm .001$ -inch height, and was made to have a .010-inch wall thickness. The inner containment jackets are also fabricated to have a .010-inch wall thickness, and each capsule nests tightly within its outer container after insertion and expansion at assembly.

The packaging method is quite simple in concept: each of three concentric gold capsules is held by a pair of stainless steel restraining discs (see Fig. 1). The lower disc is made to be the exact thickness of the capsule after it is closed and sealed; the upper disc thickness is equal to the difference between the closed capsule and the original cylinder length before closure is made.

We therefore have six types of stainless steel discs (three pairs of two discs each) to constrain the three sizes of capsules (Fig. 1.5). We designed the capsules to have a clearance of about 2 mils between the concentric gold cylinders prior to closure; therefore the capsules will easily nest inside each other before assembly. The outside diameter of all the restraining discs are precisely the same so that they can be stacked up in a column in true axial alignment.

An aluminum cylindrical guide is provided to keep these discs (nicknamed washers) in true alignment during the assembly sequence, and to keep later closure tools in precise alignment as well (see Fig. 2).

Each of the washers for a given capsule size has two small half-circle cutouts 180° apart. The washers are assembled on the gold capsules 90° out of position with each other, so that the two restraining discs can be separated with special parting tools after the capsules are loaded, but prior to swaging the lids closed (see Fig. 3). Swaging tools to form the capsule closure after the lid and solder disc are inserted are positioned by the cylindrical guide, and form the closure in four steps --

the gold cylinder is formed to a conical shape first at 30° from the vertical, then 45°, then 60°, and finally at 90° to form a flat top with a small circular depression in the center of the flat top (see Fig. 4).

Preparation of the Inner (Primary) Capsule

All assembly work is performed in a master-slave operated cell enclosure. The gold capsule(s), contained in its pair of restraining washers, is placed on the support base and the assembly positioned into the aluminum cylindrical guide. A funnel adapter which matches the cylindrical guide is placed on top; this adapter has a shoulder extending into the gold capsule to prevent loss of powder to the die assembly during the filling operation. The precise amount of actinide oxide and gold powder (-200 mesh powder) needed to form the cermet is placed into a remotely-operated mortar and pestle grinder unit, and the mixture is ground together for about 10 minutes. This powder is then transferred through the funnel assembly into the gold capsule. After brushing the residues into the cavity, the funnel adapter is removed and a die having the same bore as the gold capsule inside-diameter is placed over the gold capsule. A compaction ram is inserted into the die, and the entire assembly placed into a small hydraulic press assembly (see Fig. 5). The powder is compacted at 78,000 psig for 1 minute, the assembly is removed from the press, and the die and ram removed from the guide. A 10-mil gold lid is placed on top of the compacted cermet, and a disc of fusible gold-tin* solder placed on top of the lid. The gold capsule, still restrained in two washers, is removed from the guide tube and placed in a jig used to separate the two washers (see Fig. 6). The upper washer is removed by

*As an alternate, SIL-FOS[®] brazing alloy can be used.

application of vertical force from the slave manipulator hand, and the gold capsule containing the cermet, lid, and solder disc remains in the lower washer with the section of the gold cylinder to be swaged-closed exposed above it.

The assembly is now placed back in the cylindrical guide. The gold cylinder is closed flat by use of a hydraulic press and the sequential application of the four swaging tools. The last swage forms a smooth, flat surface, flush with the upper face of the remaining restraining washer.

The gold capsule, still held in the stainless restraining washer, is placed on a heated surface to fuse the solder residing between the capsule lid and the now flat capsule top. The fusion seal is completed in 60 to 120 seconds, as indicated by a liquid meniscus appearing in the small central depression left in the top face.

After all of the group of first-stage capsules are made in a similar manner, they are cleaned in an ultrasonic cleaning bath to remove the alpha contamination on the capsule assembly. The capsule(s), still mounted in the restraining disc, is cleaned by using dilute nitric acid, strong detergent, water, and acetone in that sequence. After drying, the first-stage capsules, now free of loose alpha activity, are carefully transferred to a lower-level-contamination manipulator-operated cell box where they are encapsulated in a second gold jacket.

Second Encapsulation

The second stage encapsulation is performed with equipment closely resembling that which is used to form the primary capsule. This cell contains a second small hydraulic press, a group of intermediate-size gold capsules held in restraining discs, second-stage swaging tools, a cylindrical guide, and the jigs necessary for separating capsule restraining washers.

Instead of forming a cermet, we now simply eject the previously formed primary capsule, lid-down, into the second gold container. A 1-mil wafer of pure copper with a central hole the size of the first capsule outside-diameter is used to protect the second capsule top-restraining-disc from being contaminated by contact with the primary capsule restraining washer. After ejection of the capsule from the primary washer, that washer is discarded along with the protective copper gasket, and the entire closure sequence outlined for the primary capsule is repeated.

After resoldering the assembly, the cleaning sequence is repeated as before; at this point the contamination level of the capsule assembly is usually less than a few thousand alpha disintegrations per minute by direct surface measurement.

Third Encapsulation

A third encapsulation identical to the second encapsulation is performed in yet another, essentially-uncontaminated, manipulator-operated cell enclosure. The sequence is performed with a third set of tools sized to match the third capsule. The product from this third encapsulation is free of measurable alpha contamination.

To bring the assembly to proper dimensions, the triply-clad cermet assembly is forced through a slightly tapered sizing-die where it emerges as a perfect right circular cylinder, meeting the diametrical tolerances to ± 0.0002 -inch (see Fig. 7). The length of the capsule is determined by the quantity of powdered gold and actinide oxide originally placed into the primary capsule. Typically, the height of the finished product can be held to ± 0.0005 -inch in length, quite adequate to meet our requirements.

Proof Testing of Capsules

In order to establish the integrity of the capsule, we have pressure tested several capsules produced during actual operations. A 63-mil hole was drilled through the bottom-center of the final assembly (into the curium-gold cermet) with a precision drill; the hole terminated in the center of the cermet. A 62-mil OD, SS-316 capillary tube was brazed into the assembly using "easy-flow 45" brazing compound after a tungsten wire was inserted into the capillary tube to keep the bore open. The capsule assembly was then installed inside a small vacuum system constructed from mini-conflat[®] flanges, with the capillary tube extending from the flange through a HIP[®] coupling. The vacuum container was connected to a helium leak detector capable of detecting 10^{-8} std cc/sec of helium leak rate, and pumped down to 10^{-5} torr vacuum through one stage of HEPA filtration. After a base vacuum was established, the spectrometer indicated a very slight helium signal. We then pressurized the capsule with helium gas in 100 psig increments. There was no indication of helium leakage from the capsule until >1900 psig helium pressure was reached. At this point catastrophic failure appeared to occur; the test was terminated and the assembly opened in a hot cell.

We found the capsule surface to have bulged into a near hemisphere on the front face, but no other significant features were noted. Subsequent tests on other capsules gave similar results. The failure pressure was >1500 psig in every case. This pressure of helium could not be reached by alpha decay for many months of storage due to the small amount of void space left in the cermet, thus the capsules were deemed satisfactory for our use.

Long-Term Storage

We have had occasion to store several capsules made by the above method in a restrained cavity for a period exceeding five years. We note there is no detectable deflection of the flat face of the capsule as determined by precision measurement of the capsule dimensions. Calculations indicate that helium pressures in excess of the test pressures used would be formed if all the helium from alpha decay had been retained. We conclude that the helium from decay has gradually diffused through the microporous plated gold structure.

An inspection of the plated gold capsule walls shows the presence of micropores in the deposited gold surface, confirming our hypothesis.

Interestingly, there is no detectable surface alpha contamination on these capsules after storage for over five years. Apparently the pore size in the plated-gold surface is fully adequate to retain all particulate contamination.

CONCLUSION

The described technique is a relatively simple method for encapsulation of highly-radioactive materials in a high-density, ductile container.

It might well be adapted for use as a containment technique for highly alpha-active neutron emitters such as ^{252}Cf where large amounts of alpha activity are inherently present, and it is desirable to use small, high-density containers to avoid degrading the spontaneous fission neutron spectrum for experimental reasons. The method is simple to perform, uses inexpensive equipment, and can be performed quite rapidly after a short learning period.

ACKNOWLEDGMENT

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FIGURE CAPTIONS

- Fig. 1. Three sizes of gold capsules assembled in restraining discs, with capsule lids.
- Fig. 1.5. The component parts required to fabricate one complete capsule, shown disassembled.
- Fig. 2. Assembly setup to eject a primary capsule into a secondary jacket (guide cylinder removed).
- Fig. 3. The tools used to remove the upper restraining disc from gold capsule.
- Fig. 4. Illustration to show how gold capsules are swaged closed in four stages.
- Fig. 5. Hydraulic press with assembled guide unit prior to capsule ejection; swaging and separation tools in foreground; tapered sizing die.
- Fig. 6. Removing an upper restraining disc from a loaded gold capsule.
- Fig. 7. A completed, triple-encapsuled, tracer package.

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CONCEPTUAL ASSEMBLY

Crimping