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EXAMINATION OF JACKETED URANIUM SLUGS FROM THE LOS ALAMOS FAST REACTOR "CLEMENTINE"

by

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ABSTRACT

Examination of clad uranium reflector slugs which failed by rupture and swelling during the first year of operation of Clementine revealed no anisotropic damage to the uranium because of burnup or thermal cycling. The cladding was found to be of high-sulphur-free-machining steel, and it is postulated that thermal stresses caused welding flaws to open enough to permit seepage and corrosive attack upon the uranium cores by the reactor coolant (mercury).

A. INTRODUCTION

The initial loading of the Los Alamos (LASL) fast reactor, popularly known as "Clementine," included in its core section a number of reflector slugs of normal uranium. At the time (March 1949) the reactor was first brought to full power,(1) the instability of uranium under thermal cycling and irradiation was beginning to receive careful attention by reactor designers. Therefore, when sticking of control rods began to be experienced about a year later, the reactor was opened and the fuel and reflector slugs were examined. Two of the latter were found to be "blistered" or swollen enough to stick in the fuel lattice cage and to require special procedure for removal. One of them proved to have a ruptured can, the other a slight swelling at one end. Because of this experience the core was modified to eliminate all uranium slugs, under the assumption that the distortions were typical of the behavior to be expected.(2,3)

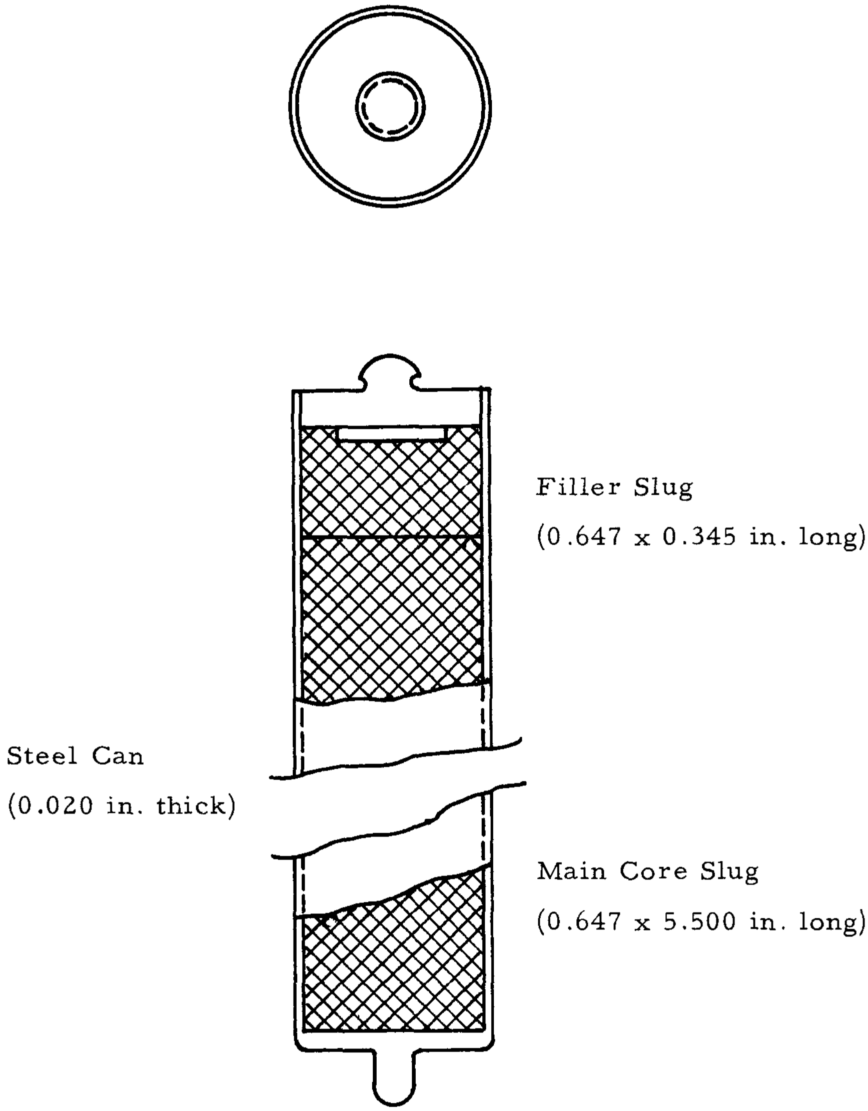
The two slugs, together with one which gave no trouble in removal, were shipped to Argonne National Laboratory for examination to determine, if possible, the cause of failure. Findings were transmitted to Los Alamos by correspondence. However, the project literature gives no record of this work, and leaves the impression that the damage is that which normally would be expected in gamma-extruded uranium.(3,4) This report, therefore, sets forth the results of the investigation from the several letters and memoranda available, and is the final record of the ANL study program.

In view of the permanent disassembly of Clementine in 1953(2) and the fact that liquid sodium-potassium alloy technology has made unlikely the design of mercury-cooled reactors in the future, the investigation is now of historic interest only.

B. METALLURGICAL AND IRRADIATION HISTORY OF SLUGS

The basic slug assembly is drawn in Figure 1, and the technical details of its fabrication, as reported in the literature,[1] are given in Appendix A. The main core was a $\frac{5}{8}$ x $5\frac{1}{2}$ in. slug augmented by a filler piece, approximately $\frac{3}{8}$ in. long, both probably of gamma-extruded uranium. The can was of mild steel, machined to 0.020-in. thin-wall dimensions from round stock. There were some uncertainties in metallurgical definition of the materials used.[3]

Figure 1. Sectional Drawing Showing Slug Assembly



In fabrication, the assembly (two-piece core, can and an end plug) was coated with a lubricant and pushed through a die to obtain tight contact between can and core. The can was then trimmed and a nonfluxed arc weld was made around the end cap. All operations were done in a helium atmosphere. Those slugs used in the reactor passed careful Zyglo and helium leak tests.

The assemblies were reported(5) to have received a total of 19,000 kwh exposure in Clementine in a fast flux of approximately 5×10^8 nv per watt. In the one year period of operation it was estimated that the reactor had been brought up to power about three times a day. Because of this the slugs received 500 to 800 thermal cycles from 25°C to approximately 150°C, in addition to fission and fast neutron damage.

The burnup due to fission is reported(3) to be 1.4×10^{19} atoms in each 580-gram uranium slug, 4% of these being U-235 atoms. This would amount to one atom in each 100,000, or 0.001% total atom burnup. The same value is obtained from the kwh and nv per watt data mentioned in the preceding paragraph if the effective fast fission cross section for U-238 is taken to be 0.3 barn. This value is not unreasonable, and in any event could not exceed 0.6 barn. Therefore, the upper limit for total atom burnup is 0.002%, with a probable value of approximately 0.001%.

C. EXAMINATION AFTER IRRADIATION

The three slugs received at ANL for examination were identified as Nos. 141, 205 and 236. Slug No. 141 was swollen and had a longitudinal split in its steel can, extending from the welded end a distance of 2¼ in. along the length. The end cap at the weld was nearly completely girdled by a circumferential crack and was very easily broken loose, after which the small uranium filler piece in the core fell out. This slug had been radiographed at LASL, and a profusion of surface bumps on the main core piece had been noted.(3) Slug No. 205 had stuck in the reactor, but was received intact. There was very slight visual evidence of a bump approximately 1½ in. from its welded end. Slug No. 236 was without detectable visual flaws. However, careful measurement of its diameter by means of a micrometer revealed that it, too, had a bump on it, somewhat closer to the welded end than No. 205, and almost as large. Length and diameter measurements are given in Table I.

Damage to Uranium Cores

After the dimensions had been measured, the jackets were carefully removed from all slugs. The 0.1-curie total activity of each was low enough to permit using a hack saw to girdle the ends of the cans and score them longitudinally by hand, using lead bricks to shield the operators. Examination of the can from the ruptured slug will be described in the next section.

TABLE I

Dimensional Measurements of Slugs after Irradiation

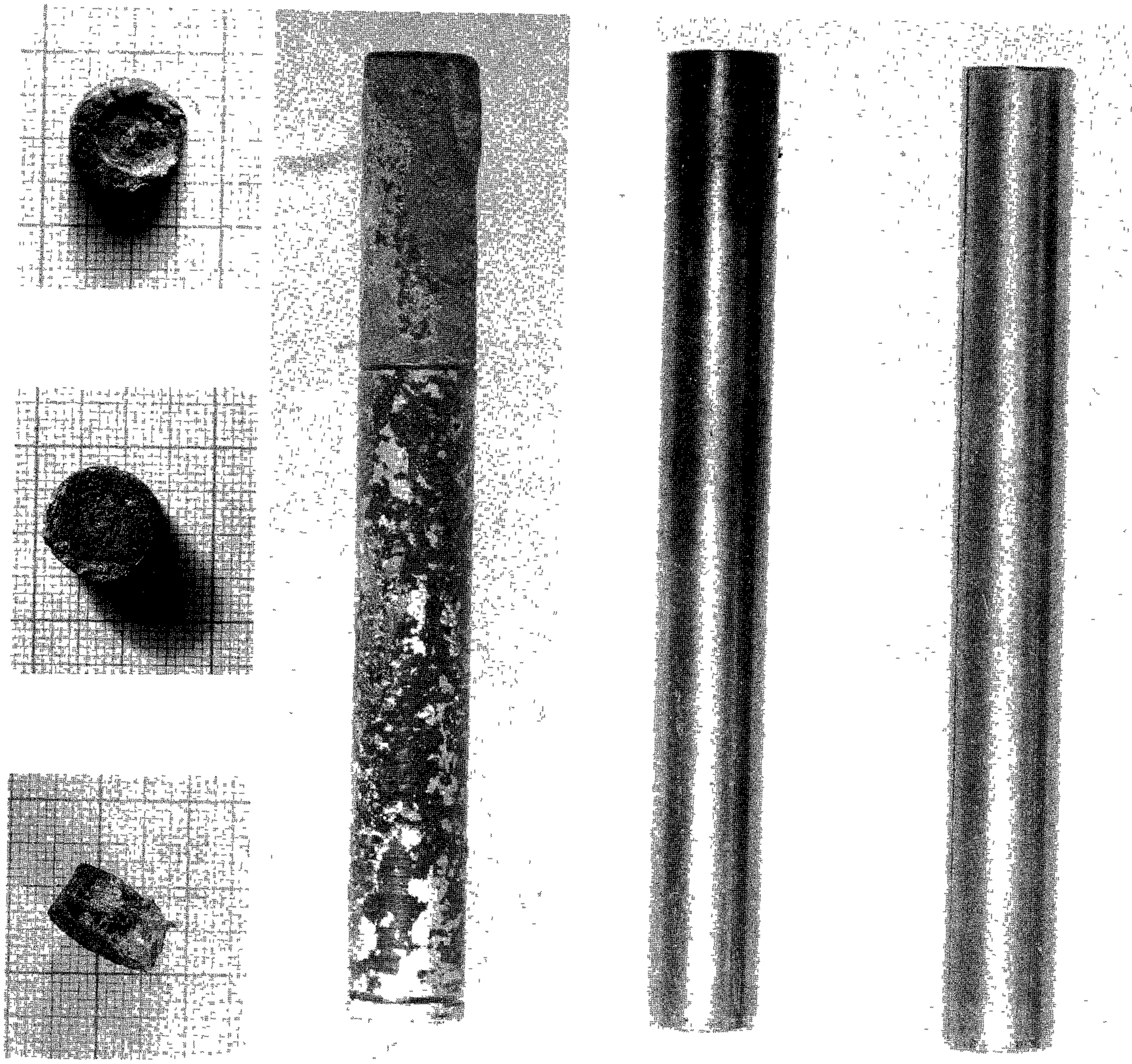
Dimension	As Received (in.)			After Removal of Can (in.)			
				Ruptured Slug 141			
	Ruptured Slug 141	Stuck Slug 205	Free Slug 236	Before Pickling	After Pickling	Stuck Slug 205	Free Slug 236
Length [a]	-	6.1012 ±0.0020	6.0743 ±0.0015	5.507 ±0.002	5.4952 ±0.0004	5.5152 ±0.0030	5.5051 ±0.0030
Diameter [b]							
Average	-	0.6878 ±0.0002	0.6875 ±0.0004	0.645 to 0.656	0.6412 ±0.0004(7)	0.6466 ±0.0002	0.6471 ±0.0002
At Bumps	-	0.696 to 0.698(3)	0.689 to 0.695(5)	0.657 to 0.660(2)	0.645 to 0.650(3)	0.648 to 0.649(4)	0.648 to 0.652(4)

[a] Averages based on 5 measurements.
[b] Based on 10 to 14 measurements, except as indicated otherwise in parentheses.

The overall appearances of the decanned slugs and of the small uranium filler plug from the ruptured slug are shown in Figure 2. The appearances of surface details are seen better in Figure 3. The core of the failed slug was covered with bumps and erosion. It had been silver-plated before assembly, whereas the other two cores were unplated. In Figures 3-b and 3-c may be seen the small bumps on the surfaces of the latter slugs which were associated with the out-of-roundness of their respective cans, noted above. Careful measurements (Table I) showed these irregularities to be the only ones present on the surfaces of the cores. They had the same general appearance as the smaller surface bumps on the ruptured slug, except for the absence of silver plating, but equipment was not available whereby they could be studied more critically.

Two regions of damage on the ruptured slug core appeared characteristically different. 1) The end in the vicinity of the jacket rupture had no large bumps, was heavily eroded and had a dull gray, oxidized appearance (Figure 2-b). The small filler plug had a similar appearance. It was also noted that, on this piece, the surface closest to the weld had considerable more erosion than the surface in contact with the main core slug (Figure 2-a). 2) At positions remote from the rupture the damage took the form of many discrete bumps between which could be seen tool marks originally left on the core surface by the machining operation.

After photography and dimensional measurements, the bumped area of the ruptured slug core was scraped to obtain a sample for chemical analysis. The bumps were seen to be composed of gray material which readily ignited when disturbed, giving off pyrophoric flashes. Spectroanalysis of the crud revealed it to be primarily uranium and silver, but



9050, 9052, 9053
a)

10221
b)

10337
c)

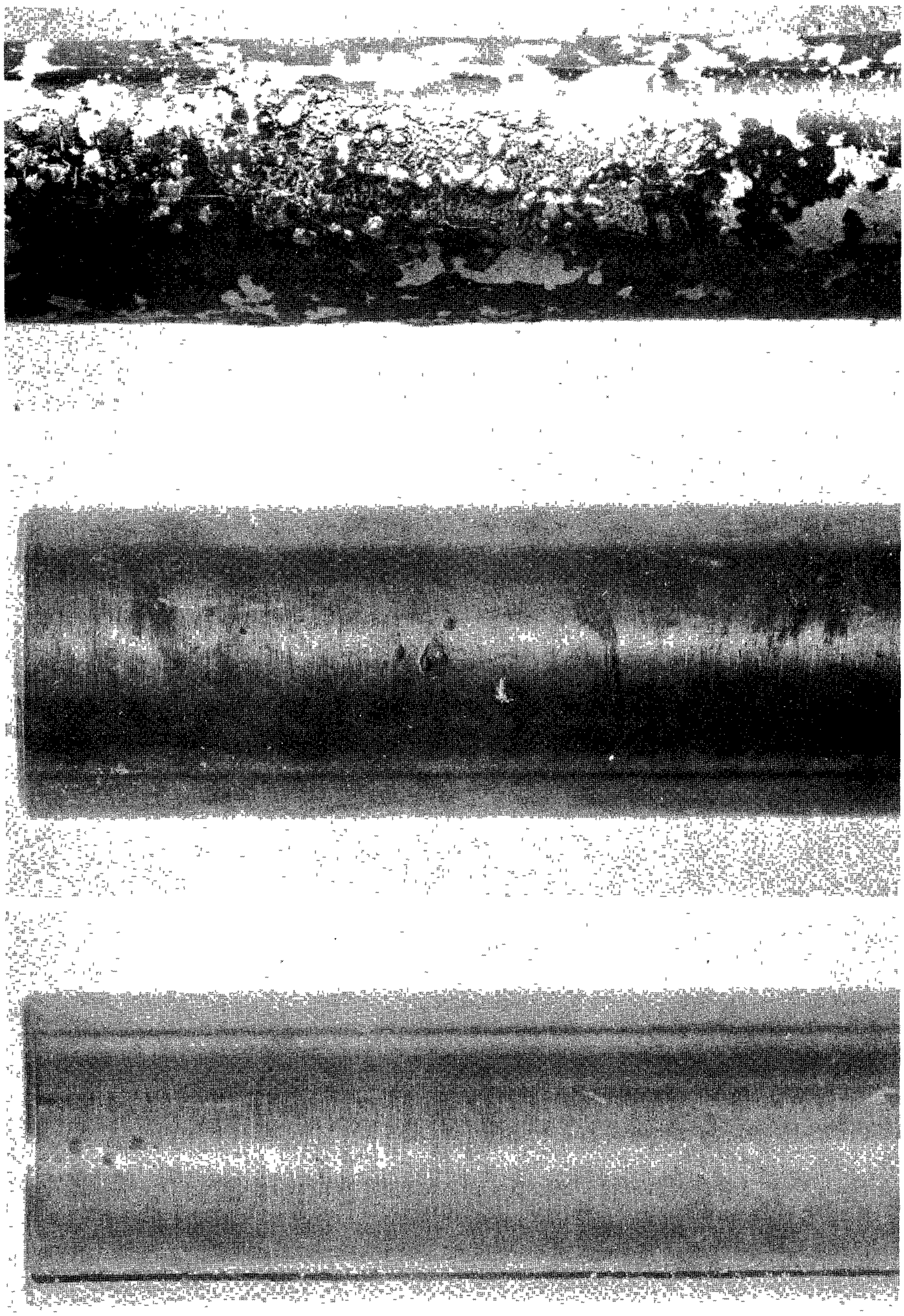
10342
d)

Figure 2. Appearances of Uranium Slugs after Removal of Cans (1X).
a) Three views of end filler from Slug 141 (ruptured).
b), c), d) Main cylinders from Slugs 141, 205 and 236, respectively.

a)

b)

c)

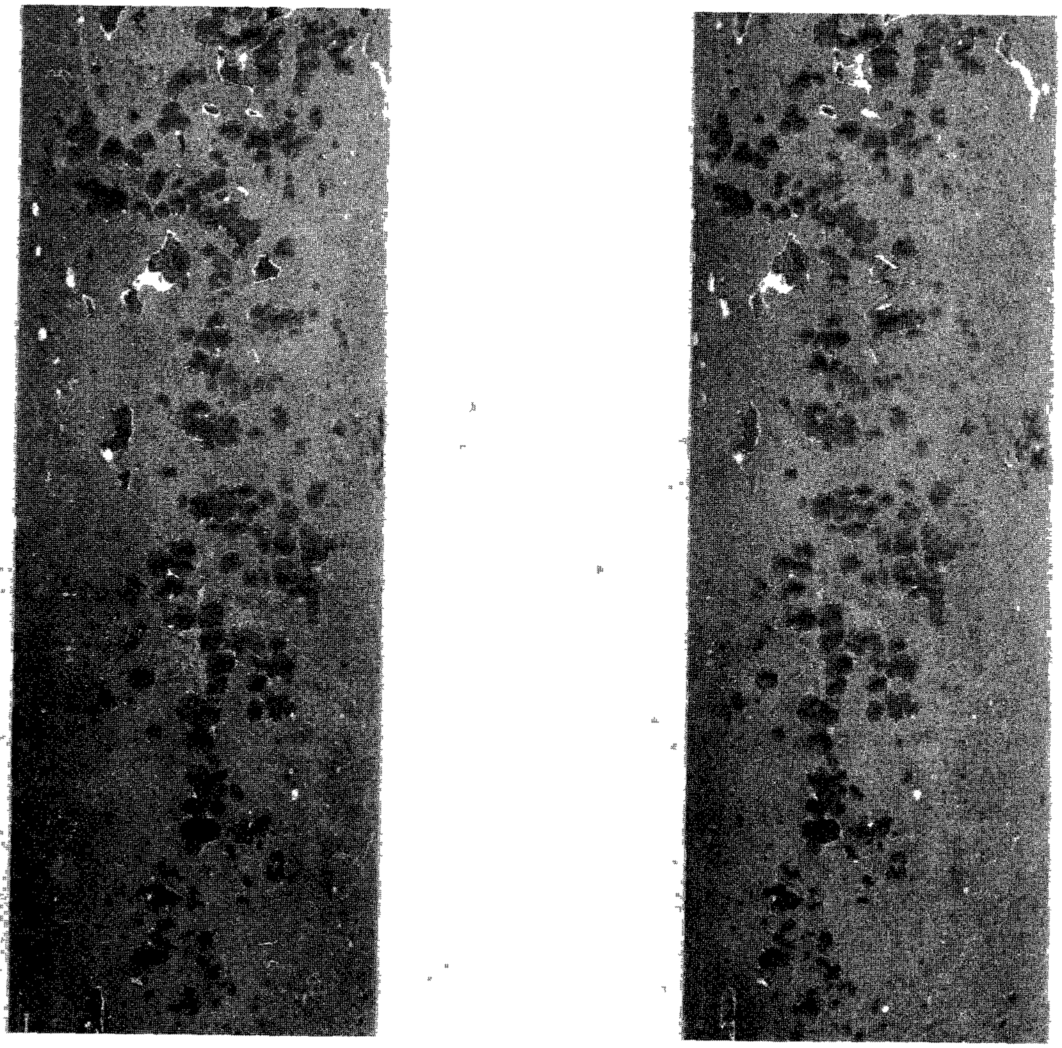


10218, 10339, 10341 ~3X

Figure 3. Enlarged Views of Stripped Slugs. a) Profuse bumping of ruptured Slug 141. b) c) The only irregularities noted on Slugs 205 and 236.

there were, in addition, a moderate content of barium and silicon, lesser amounts of aluminum, calcium, iron, magnesium, manganese, lead and titanium, and a very small amount of chromium. Surprisingly, only a trace of mercury was reported, although droplets of the liquid metal were present in the scrapings.

The core of the ruptured slug was next pickled in 50% aqueous nitric acid for a total of ten minutes. The silver plating and raised bumps were attacked and removed quite rapidly, leaving a matte metallic surface in which pits occupied the positions of all the bumps. Pickling was stopped before the reaction went to an end. Figure 4 shows the appearance of the slug after it was washed and dried. The stereo permits seeing in three dimensions the pits and residual nonmetallic gray material contained by them. A cross section through one of the pits is seen in Figure 7. The grain structure of the uranium is coarse and typical of gamma extruded



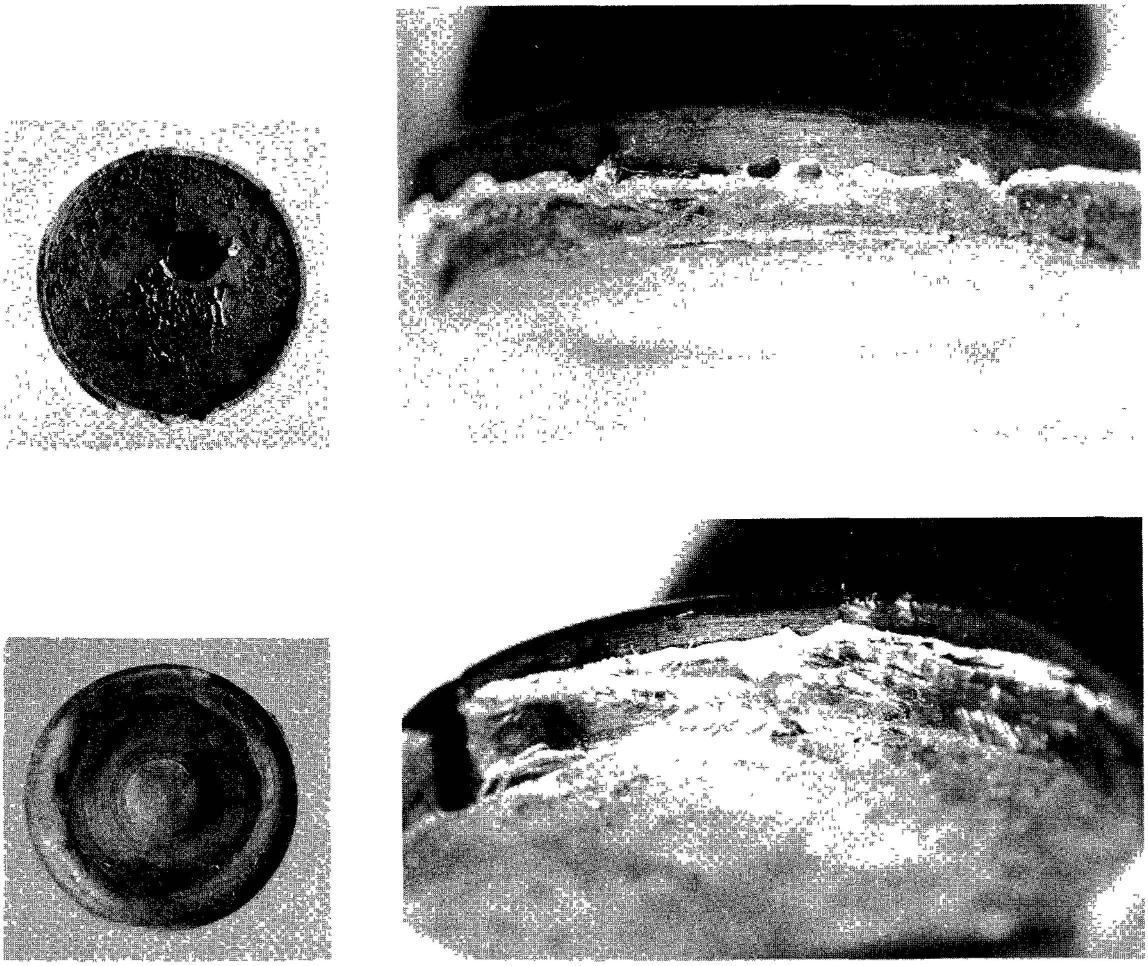
10245 10244 ~3X

Figure 4. Stereomacrograph of the Core of Ruptured Slug 141 after Acid Pickling.

material. It is of interest to note that the corrosive attack which formed this pit was transgranular, with preferential action in certain definite directions in the grains.

Data from Ruptured Jacket

Steel in all cans and end plugs proved to be ferromagnetic. The appearance of the end cap removed from the ruptured slug is recorded in Figure 5. Its inner surface was covered with a coating of crud, some of it black and vitreous, and some similar to that on the filler plug. The glassy black material proved to be insoluble in aqua regia, and analysis indicated that it contained appreciable amounts of barium and manganese.



9051, 9054 2X 9048, 9049 5X

a) b)

Figure 5. Views of End Cap from Ruptured Slug 141. a) As removed from slug, showing inner vitreous deposit, and small blowhole in outer weld. b) After peeling can back to weld line, showing blowholes and "flakes" along same.

The welded closure of the end (Figure 5-a) seemed visually sound, except for a hint of a surface pit where the arc had been struck. However, it was prepared for further study by carefully peeling back the small ring of attached can material so that the line of fusion all around the end cap could be examined more critically for points of leakage. Two blowholes were found adjacent to each other; one of these extended through the thickness of the can (0.020 in.) and had apparently formed a leak from outside to inside. The inner surface of the hole was discolored (Figure 5-b) and the discoloration continued inside along the side of the end plug toward the uranium. Several flakes in the weld were also observed, one of which (Figure 5-b) was discolored in such a way as to suggest leakage.

Examination of the steel can in the vicinity of the rupture showed a brittle type of fracture in this material. It was impossible to flatten out a cut section of the jacket without breaking it along the rolling direction. Hardness of the material was Rockwell 15T 88.8 to 89.9 (R_B 86-90 converted), which is considerably higher than would be expected in annealed mild steel. However, metallography confirmed its low carbon content (approximately 0.20%), as may be seen in the etched structure (Figure 6). The presence of many manganese sulphide inclusions in the structure labeled it as free-machining steel, and an analyzed sulphur content of 0.175% confirmed this point.

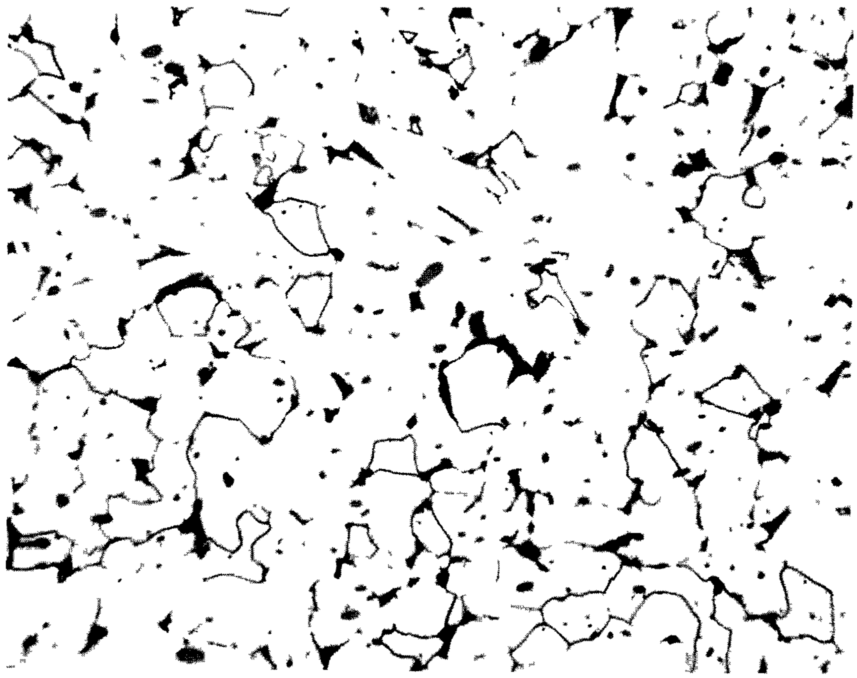
D. DISCUSSION

The effects of neutron irradiation and of thermal cycling upon coarse-grained uranium, such as that produced by casting or gamma extrusion, have become well known.[6-9] If the grains are randomly oriented, and if irradiation or cycling is continued, gross distortion and bumping of the surface occurs and characteristic changes in the microstructure will take place. Fine-grained uranium, such as that obtained by sharp quenching from the beta phase, has a surface stability many times greater than gamma-extruded uranium. It is, therefore, much more suitable for use in reactor slugs.

The LASL decision to eliminate gamma-extruded uranium slugs from Clementine is, therefore, fully justified by general experience with this material. However, the full weight of the ANL investigation of the failed slugs seems to support the viewpoint that the same trouble would probably have been experienced if the uranium slugs had been made of fine-grained stable material, other variables remaining the same. Substantiation of this viewpoint is as follows:

1. The rupture and sticking of the slugs in their reactor channels was very evidently due to the buildup of the reaction products of mercury and uranium in localized bumps. As the slugs were all leak tested before their irradiation, it follows that whatever caused or permitted the entry

10078 As polished 300X

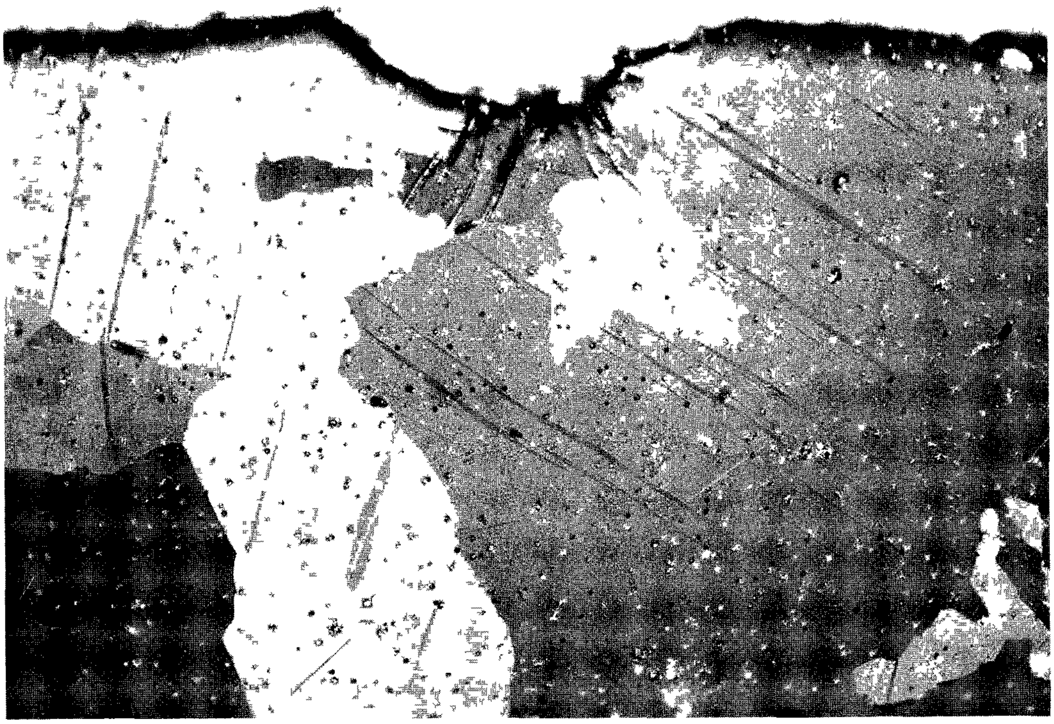


10077 2% Nital Etch 300X

Figure 6. Photomicrographs of Steel Can from Ruptured Slug 141

of the liquid metal coolant is responsible for the failure of the slugs. Only a dynamic factor could induce the initial perforation.

2. Anisotropic deformation of uranium is a dynamic factor, under proper conditions of irradiation and/or thermal cycling, which might create localized stresses in a slug jacket. However, in the present case none of the bumps found on the cores was a protrusion of uranium metal. The complete absence of generally roughened metal on the surfaces of any of the cores (Figures 2, 3 and 4) is proof that anisotropic radiation and thermal cycling damage had not occurred. Uniformity of the dimensional measurements (Table I) reinforces this point. In addition, the microstructure of the uranium (Figure 7) is not characteristic of the metal that has undergone cold work of any appreciable degree. Work done at ANL[7,9] makes it very doubtful that any amount of thermal cycling from room temperature to 150°C could produce appreciable surface roughening. Also, the threshhold for noticeable roughening due to irradiation growth of surface grains is probably an order of magnitude higher than the burnup of these specimens.[6,8] These considerations, therefore, seem to indicate that anisotropic deformation of the uranium core material was not responsible for the slug failures.



14248 Polarized Dark Field 100X

Figure 7. Photomicrograph of Uranium Core of Ruptured Slug 141, Showing Gamma-extruded Structure and a Surface Pit

3. An alternative mechanism, which could account for perforation of the slug jacket, is proposed. Uranium has a bulk expansion coefficient some 28% greater than that of iron in the temperature range being considered. Heating to 150°C from room temperature would create a tensile stress of approximately 12,000 psi in the steel cladding if the core and clad were initially unstressed and in good contact. Free-machining steels are notoriously deficient in transverse properties, have bad welding characteristics and are subject to flaws when joined by fusion.(10) Stringers of sulphide inclusions act as stress raisers, and the regions around blowholes and microcracks (flakes), such as are shown in Figure 5, are already highly stressed due to cooling after the welding operation. Irradiation embrittlement and loss of ductility are further liabilities. When all these factors are weighed it seems quite likely that the mild thermal cycling experienced by the slugs could have added enough of an overstress to crack the high-sulphur steel thin wall and permit initial seepage of mercury.

The presence of the black vitreous material within the ruptured slug remains an anomaly. Barium, silicon, aluminum, calcium, iron, magnesium, manganese, lead, titanium and chromium in the spectrographic analysis of crud lead one to suspect that welding fluxes were used in fabrication of the slugs and/or that corrosion inhibitors were present in the mercury. Direct inquiry about these possibilities(11) removes them from consideration. The possibility remains that sizing lubricant contaminated only the ruptured slug assembly before welding. The unruptured slugs did not have within them any trace of the vitreous black scale found in the ruptured slug. Another possibility is that the circulating mercury picked up contaminants such as residues from welding fluxes used in fabricating the reactor vessel. However, such transport deposits are usually expected in the cooler portions of the coolant loop. Moreover, there was no trace of external deposit on any of the slugs. At any rate, it is not likely that this crud had a primary bearing on the failures.

As a last comment, it is of interest to note that the grain size of the core uranium was approximately $\frac{1}{32}$ in., and that the pits remaining after pickling of the ruptured slug core were of the same approximate size. Corrosive attack by the mercury was apparently preferential to an orientation factor in the grains of uranium and proceeded most rapidly in those that were favorably exposed. General attack of the uranium surface was almost completely absent where the mercury was in static contact, and does appear only in the vicinity of the gross rupture where dynamic erosion probably was proceeding.

E. CONCLUSIONS

The failures observed in the uranium reflector slugs initially installed in the Los Alamos Fast Reactor, Clementine, are attributed to the use of high-sulphur free-machining steel as a cladding material. The fact that coarse-grained uranium was used in the core would probably have caused trouble after irradiation to burnups an order of magnitude or so higher. However, presence of the anisotropic damage characteristic of coarse-grained uranium under critical conditions of radiation and thermal cycling is not observed in the failed slugs.

F. ACKNOWLEDGMENTS

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APPENDIX A

Fabrication of Reflector Slugs

Reflector rods were fabricated from gamma-extruded normal uranium, machined to size and canned in a mild steel jacket to allow handling and prevent corrosion from contact with the mercury coolant. The following account of procedure is abstracted from LA-1679.(1)

Dimensions of the plated reflector slugs are 0.647 ± 0.002 in. in diameter and 5.500 ± 0.005 in. long. Canning operations were performed in a helium atmosphere.

The first step was to place the slug in a machined can of 1020 steel having an inner diameter of 0.652 ± 0.001 in. and 0.020-in. wall thickness. These cans have a bottom thickness of 0.062 in. and are made with a centering pin $\frac{1}{8}$ in. in diameter and $\frac{3}{16}$ in. long. All steel cans had been annealed in a hydrogen atmosphere after machining to remove strains. A wafer of normal uranium, 0.345 in. thick, was used to cover the reflector slug. This uranium wafer has a recess $\frac{1}{16}$ in. deep by $\frac{3}{8}$ in. in diameter in which fission gases could accumulate. The final steel cap for sealing the can was then placed on top of the wafer. The assembled rod was painted with a drawing compound and pressed through a sizing die to give a diameter of 0.686 in. The sized can was then trimmed to proper length and arc-welded in the helium atmosphere.

After canning, the rods were tested for leaks in the welds by two methods. 1) The canned rods were soaked in Zyglo (fluorescent) oil at 150 psi for 3 days, then removed, cleaned, and examined under ultraviolet light for oil seepage; this test was repeated four times on each can. 2) The cans were soaked in helium at 90 psi and then placed in a vacuum chamber connected to a mass spectrometer leak detector. The Zyglo method is reported to detect holes 3×10^{-10} cm^2, whereas the mass spectrometer method can detect 1×10^{-11} cm^2 holes.