December 1, 1969

To: Distribution

From: G. L. Ragan

Subject: Procedures for Examination and Analysis of U-Capsules Irradiated in MSRE for the Eta Experiment


Following the design described in Ref. 1, six capsules were made, filled, and sealed this past June — in record time, thanks to the excellent cooperation and extra effort of everyone involved. Four have been irradiated in the MSRE during its subsequent operation and two are being held as controls. Present plans call for leaving the capsules in the MSRE during the next short run (a week or so at power) and removing them about the middle of December.

I summarize below my understanding, based on our earlier discussions, of procedures that will yield the desired information from these irradiated capsules and the unirradiated comparison materials. Should my understanding or recollection of our hastily made plans differ substantially from yours, or should you see a better way to accomplish our objectives, please let me know.

1. Disposition of Materials on Hand (C. H. Gabbard)

   We will need to know the initial concentration of total uranium (gU/g salt) in the two types of sample salt prepared by J. C. Mailen. The bottle of each type salt that contains the smallest amount (roughly 8 g of one and 16 g of the other) should be sent to Mailen for this determination. He will also use this material to prepare electroplated sources of each type for U-isotopic analysis by A. E. Cameron.

   The remaining four bottles of salt, and the two unirradiated capsules, will be sent to G. Goldberg when he is ready for them.

2. Capsule Opening (E. M. King)

   Remove the two short and two long capsules from the experimental assembly, noting the identifying mark etched on each capsule bottom and its position in the assembly. The order should be (from top downward) S2, L4, S1, L2. These four capsules plus two unirradiated capsules (S3 and L3) are to be opened and contents recovered. Special care is required to avoid cross-contamination between capsules or contamination from other sources — especially from MSRE fuel that may adhere to the outside of the capsules. Contaminations by U or Li are especially detrimental.
The following procedure is based on our earlier discussions. Grind through
the outer Mo tube near the top of the capsule and remove the top cap.
Position the cut so as to facilitate alignment between the capsule and the
special funnel (made by you) that couples to the reaction vessel (supplied
by G. Goldberg). Note capsule number and corresponding vessel number (or
mark vessel to correspond to capsule number). A separate vessel is used
for each capsule, and to avoid cross contamination, a funnel will not be
reused. Melt salt (mp = 525°C) from capsule into vessel. Do not exceed
600°C, lest the indication of the irradiation-temperature monitors (SiC,
in capsule central cavity) be invalidated. If higher temperatures are
required, we must first remove the monitors from the central cavity (as
described below) and then do the salt melting. Seal the vessel and send
it to G. Goldberg.

Unscrew threaded plug from central cavity and remove it. Screw on, in its
place, the metal carrier whose number matches that etched on the capsule
bottom; invert the capsule to transfer monitors to carrier; remove carrier,
cap it, and send it to Wallace Harvey.

The metal carrier has not yet been designed, but the following is suggested.
Choose a suitable metal rod (say, 1/2-in.-hex brass, 6 in. long). Bore a
#29 (0.136 in. diameter) hole on axis, 5 3/4 in. deep; strongly chamfer
the entrance end of the hole to guide the pieces in; turn down the outside of
this end and thread it the same as the threaded plug. (Instead of
threading, an alternative means for aligning and securing parts may be
guessed.) Put a plug of cotton, as shock absorber, in the bottom of this
hole; leave sufficient free length (2 1/2 in. for short, 5 1/2 in. for
long capsules) to accommodate all monitor pieces, even if the small wires
fall out of their graphite holders. Six carriers, with suitable caps, will
be required. Mark the carriers to correspond to the capsules: S1, S2,
S3, L2, L3 and L4.

3. Flux Monitor Measurements (R. W. Harvey)

Remove the small monitor pieces from each carrier, taking care to preserve
their sequential order. Place each piece in a suitable small container
appropriately marked with its carrier number and its sequence number
(order of removal from carrier, starting at the bottom of the enclosed
figure). Send the SiC pieces to C. R. Kennedy for evaluation. Retain the
graphite pieces until we see if anyone has a use for them. Use the metal
pieces for your flux monitoring measurements.

Note that two sets of monitors are from unirradiated capsules. These need
not be examined unless you see an advantage in doing so. I have, in
addition, one set (for a long capsule) that was never put into a capsule.

Our interest is in variation with height (i.e., from monitor to monitor
along the string of capsules) of (a) the "thermal" flux (corresponding,
in this case, to over 600°C), (b) the "epithermal" flux, and (c) the "fast"
flux. (Calculated fluxes are given by B. E. Prince in MSRP Semiann. Progr.
Rept. Aug. 31, 1967, ORNL-4191, pp. 50-58.) We will need, for each monitored activity, the number of disintegrations/sec per mg of monitor for the following activities: $^{110}$Ag from the Cu-Ag alloy pellets; $^{60}$Co, $^{58}$Co, and $^{54}$Mn from the 302-SS wires; any others you find relevant.

Interpretation of these data will be considered in detail later on — after the irradiation is completed and the power history is available.

4. Temperature Monitor Measurements (C. R. Kennedy)

Four pairs of SiC monitors (rods 0.1 in. diameter by 0.5 in. long) have been irradiated (in capsules $S_1$, $S_2$, $L_2$, and $L_4$) at temperatures estimated to be in the 680°C to 750°C range. Annealing curves, to determine irradiation temperatures, are needed for these. Available for comparison, if desired, are two pairs of SiC monitors from unirradiated capsules and an additional pair that was never put into a capsule. As you may recall, we anticipate that length changes on annealing may be small and difficult to measure. (I made an order-of-magnitude estimate that we can expect about 0.3 mil change in length on annealing.)

5. Chemistry (G. Goldberg)

We will need ten reaction vessels — send six to E. M. King for the encapsulated salt samples ($S_1$, $S_2$, $S_3$, $L_2$, $L_3$, $L_4$) and keep four for the comparison samples ($S_4$, $S_5$, $L_1$, $L_5$) being held by C. Gabbard. The vessels should be clearly marked — preferably with the above designations ($S_1$ through $S_5$ and $L_1$ through $L_5$). Each short capsule will contain about 8 cc (25 g) of NaF-ZrF$_2$UF$_4$ salt, long capsules about 16 cc (50 g) of salt. Gabbard will send you two sealed bottles, each with about 25 g of salt, for loading into vessels $S_4$ and $S_5$; also, two with about 50 g each for $L_1$ and $L_5$. Fluorination of each vessel will yield about 1 g U, separated from the NaF-ZrF carrier salt and deposited (as UF$_6$) on a NaF trap. These traps are to be sent to L. A. Smith for isotopic analysis. Each trap should be marked with the corresponding reaction vessel number.

The salt residue from each reaction vessel will be recovered. Residues from all irradiated capsules ($S_1$, $S_2$, $L_2$, $L_4$) and one comparison sample of each type ($S_3$ and $L_3$) will be analyzed; the other four will be kept in reserve. Each different residue will be analyzed as follows:

(a) A portion will be analyzed by optical spectrometry to measure the amount of Li present (spectrographic analyses run for J. C. Mailen on the initial material showed 2.67 and 2.70 ppm).

(b) One or more Pu sources (electroplated on wires) will be prepared and sent to A. E. Cameron for Pu isotopic analysis — needed in interpreting the α-count mentioned next.
(c) One or more small portions will be taken and analyzed by standard α-counting techniques to determine g Pu/g salt.

(d) The bulk of the residue will be analyzed by coulometric titration techniques to determine g Pu/g salt.

(e) Whatever auxiliary or additional steps you feel we need to determine g Pu/g salt with the desired accuracy of 0.5 to 1% (e.g., isotopic dilution or $^{238}$Pu spiking, etc.).

6. Solid-Source Mass Spectrometry (A. E. Cameron)

Analyses of the two electroplated U sources of Section 1 are needed to establish the composition of our starting material. Analyses of the electroplated Pu sources (5(b) above) are needed in interpreting the Pu α-counting data.

A small solid source from each gaseous sample will be supplied by L. A. Smith, following procedures previously worked out. This U-isotopic analysis is needed especially for the isotopic ratio of $^{234}$U to $^{233}$U (involved in our gamma equation) and fraction of total U that is $^{238}$U (needed, with Pu assay and initial concentration of U, for determining $S_8$).

7. Gaseous-Source Mass Spectrometry (L. A. Smith)

About $1 \text{ g U}$ (as UF$_6$) will be recovered from each of the ten NaF traps supplied by G. Goldberg. A small solid sample of each will be prepared, following procedures developed for the first set of $^{233}$U samples, and sent to A. E. Cameron for analysis.

The four samples from irradiated capsules represent "final" samples, in our earlier nomenclature. Four of the six unirradiated samples can be used as "initial" samples (paired with the "final" ones) and two are available as "conditioning" and equipment-check materials. The following comparisons are suggested:

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<th>Initial vs Final</th>
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<tbody>
<tr>
<td>S4 vs S1</td>
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<tr>
<td>S3 vs S2</td>
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<tr>
<td>L3 vs L2</td>
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<td>L1 vs L4</td>
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<tr>
<td>S5 conditioning</td>
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<td>L5 conditioning</td>
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Just as in the case of the $^{233}$U fuel samples now being analyzed, we are primarily interested in analyses for $^{233}$U and $^{234}$U, but want (with less stringent requirements on precision) analyses for $^{235}$U and $^{236}$U.

GLR: mw

Attachment: One figure