To: M. W. Rosenthal

From: J. R. Engel

Subject: Apparent Holdup of $^{235}$U Fuel in MSRE

The uranium recovery from the $^{235}$U fuel charge in the MSRE reported by Lindauer\(^1\) and a comparison with the anticipated recovery\(^2\) suggest that as much as 4.4 kg of uranium may have been held up somewhere in the MSRE system. This apparent holdup is in addition to the 1.935 kg of $^{233}$U shown to have been retained and mixed in with the $^{233}$U fuel charge.

The purposes of this memo are 1) to eliminate some parts of the MSRE system from consideration as possible repositories for the uranium, 2) to suggest other parts of the system, and 3) to propose an expedient method for checking the suspected areas for the presence of uranium.

**Improbable Locations**

The operating experience with the $^{235}$U fuel charge allows us to eliminate substantial parts of the MSRE as potential sites for holdup of the "missing" uranium. These parts and the reasons for their elimination are given below.

**Offgas System**

There have been reports from time to time of uranium and/or fuel salt appearing in the offgas line from the fuel pump. However, the loss of 4.4 kg of U would have had pronounced effects that were never observed.

If the uranium had escaped as an independent component (in any chemical form), the result would have been a gradual decline in the fuel salt uranium concentration by ~2%. This would have produced an

---


unexplained reactivity loss of about 0.4% δk/k; no such loss was observed. In addition, such a decline would have been revealed by the frequent chemical analyses that were performed. The analytical results were in excellent agreement with expected values, thereby eliminating this as a loss mechanism.

If the uranium had escaped as fuel salt at the operating uranium concentration, a salt loss of 1.5 ft$^3$ would be required. There was no evidence from the various loop filling operations that the salt inventory was decreasing at all. (A loss of 1.5 ft$^3$ would have made it nearly impossible to fill the fuel loop to the operating level.)

Fuel Loop

Any gradual losses of uranium to places within the rest of the fuel loop would have been detected by the same methods that would show losses to the offgas system, i.e., reactivity effects and chemical analyses. In support of these data, we have the results from graphite samples that show almost no uranium deposition. We conclude, therefore, that we had not lost a significant amount of uranium up to the end of Run 14. Since the $^{235}$U fuel salt was not put back into the loop after Run 14, the primary system can probably be eliminated from consideration.

Drain Tanks

Some of the $^{235}$U fuel charge was left in the drain tanks when the main charge was transferred to the fuel storage tank for processing. The actual amount was measured by an isotope-dilution experiment when ~0.9 kg of $^{238}$U was added to the system, prior to the $^{235}$U critical experiment. The amount found (1.935 kg) was in good agreement with our expectations, so no extra uranium was left in the drain tanks — at least not in a form that could mix readily with the salt. Even if the uranium had been left in some insoluble form (no reasonable mechanism is available to produce and retain such a material,) it would probably have undergone some isotopic exchange with the $^{233}$U in the fuel salt during the year of operation with $^{233}$U. The results of the "alpha" experiment for $^{233}$U suggest the possibility of contamination of some of the later samples with a material whose composition was similar to that of the $^{235}$U fuel. However, the scatter in the values was more suggestive of sample contamination than of systematic contamination of the entire $^{233}$U fuel charge. Even if the cause was an isotopic dilution of the entire charge, only ~35 g of uranium would have been required to produce the observed effects. Since 4400 g are to be accounted for, the likelihood that this material is in the drain tanks must be assigned a very low probability.

Possible Locations

Since the reactor system has, for all practical purposes, been eliminated as a repository for the "missing" material, it seems reasonable to

---

assume that the uranium is somewhere between the line leaving the drain-tank cell (L-110) and the NaF absorbers that were sent to Goodyear Atomic Corporation for product recovery. However, some parts of the processing plant must also be regarded as highly improbable sites for uranium hideout. The fluorination process reduced the uranium concentration in the salt in the storage tank to ~27 ppm and it is not likely that an insoluble uranium form could persist under the oxidizing conditions that prevailed during fluorination. The caustic scrubber (actually downstream of the NaF absorbers) is presumed to have collected essentially no uranium. This leaves only two major components in which uranium might conceivably remain.

Salt Filter

The first component in the line from the drain tanks that appears to have a finite probability of containing uranium is the salt filter. If the excess Zr used in reducing the processed flush salt was deposited on the filter as the flush salt was returned to the fuel flush tank, some uranium could have been reduced and precipitated from the fuel salt as it was transferred to the storage tank for processing. Uranium analyses were obtained for two fuel salt samples removed from the storage tank prior to the start of fluorination. The uranium concentrations were 4.306 and 4.293 wt.%, compared to 4.512 wt.% in the fuel salt at the end of Run 14, a decrease of 4.7% in the concentration. Most of this decrease can be accounted for. Filter runback from the flush salt transfers should have diluted the fuel salt by ~0.84% and a further dilution by 2.65% was produced by the deliberate addition of 129.9 kg of carrier salt in May 1968. Thus, the expected dilution was at least 3.49%. If, in addition, some flush salt was still in the storage tank when the fuel salt was introduced, a further dilution could have occurred. However, the final flush salt inventory does not support the existence of a flush salt heel in the storage tank. We must also assign some uncertainty to the uranium analyses on the storage tank samples. Thus, while we can account for most of the apparent dilution of the fuel salt, we cannot, with certainty, claim that all the uranium that left the drain tanks arrived at the storage tank.

NaF Trap

The uranium processing scheme included a high-temperature (~750°F) NaF trap to remove fission- and corrosion-product fluorides from the UF₆ stream. This trap was not expected to retain any uranium under the operating conditions that were maintained. However, a uranium-sodium fluoride complex can be formed (in the absence of excess F₂) that would be retained and we have no direct proof that no uranium was held up on this particular trap.

Uranium Detection

Since we cannot be certain that no uranium was left either on the salt filter or on the hot NaF trap, an examination of these two objects appears to be in order. Removal of these components, followed by dissolution and "recovery" of the uranium would be an expensive procedure that might lead to negative results. A more desirable approach might be
to examine the subject components nondestructively to obtain at least a qualitative indication of the presence of uranium.

Nondestructive methods, notably neutron interrogation and photon interrogation, have been developed for assaying the fissile-material content of objects. Of the two, neutron interrogation appears to be the most readily adaptable to this particular problem. The procedure involves bombardment of the uranium-bearing object with neutrons from an external source followed by detection of delayed neutrons with the external source removed (or turned off). Under controlled conditions with a carefully calibrated system this approach is capable of high precision (~1%) and high sensitivity (a few mg of fission material). Thus, a reasonable go, no-go indication should be attainable at the MBE with only a moderate effort.

The requirements for using neutron interrogation to look for uranium in the filter and the NaF trap are an isotopic neutron source (of the order of $10^7$ n/sec) and a reasonably efficient neutron detector. The source must be capable of rapid withdrawal (within ~5 sec) into a shield while delayed neutrons are being sought. It must also be provided with a moderator while it is in active use. A small Cf source could be borrowed for this purpose. The neutron detector should have a high efficiency in a significant gamma background. A boron-lined chamber with an efficiency of 15 c/sec/nv is available to be borrowed.

The only remaining requirements are the hardware for positioning the source and detector and neutron-moderating and shielding materials. Two vertical 6-in. pipes were installed immediately adjacent to the filter for storage of spent filter elements. If these were filled with polyethylene blocks containing appropriately located vertical holes, they would be almost ideal holders for the source and detector. Borated polyethylene could be used in the upper parts of the tubes to provide shielding for the chamber or source while the other is in use. A similar, portable assembly could easily be fabricated for use near the NaF trap. This second assembly could also be used for a rough calibration of the system.

A number of people at the Laboratory have indicated a willingness to provide assistance in the form of equipment and/or technical expertise in the performance of a "uranium assay" experiment. With reasonable diligence and luck, we should be able to obtain useful information within 2 to 3 weeks after the start of such an experiment. If negative answers are obtained for both the filter and the NaF trap, additional effort will be required to examine the more improbable "hideout" locations.

J. R. Engel

J. R. Engel

JRE:al
Distribution:

S. E. Beall
R. B. Briggs
V. A. DeCarlo
D. E. Ferguson
R. H. Guymon
P. N. Haubenreich
R. B. Lindauer
D. W. Magnuson
A. M. Perry
D. Roux
J. E. Strain
R. E. Thoma
R. H. Chapman