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PROGRESS REPORT ON THE Th-232-U-233 IRRADIATION PROGRAM AT THE MATERIALS TESTING REACTOR

Robert G. Nisle and Eugene F. Aber

PHILLIPS
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ATOMIC ENERGY DIVISION

NATIONAL REACTOR TESTING STATION

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Progress Report on the Th-232-U-233 Irradiation Program
at the Materials Testing Reactor

Robert G. Nisle and Eugene F. Aber

Summary

Four thorium-uranium samples as follows were irradiated in the MTR for three cycles:

One containing 61.58 grams Th-232

One containing 64.47 grams Th-232 and 0.6487 grams U-233

One containing 63.75 grams Th-232 and 0.8200 grams U-233

One containing 62.255 grams Th-232 and 1.0016 grams U-233

Reactivity measurements were made at two times about two weeks apart between irradiation periods. The reactivities were extrapolated to $t = 0$, the time the samples were removed from the irradiating flux, and to $t = \infty$, i.e., at long decay times compound to the Pa-233 half-life. Such calculated reactivities are a rough indication of the U-233 and Pa-233 content of the samples. The results indicate that the U-233 content is changing least in the sample that originally contained 1.3% U-233. These samples are presently scheduled for ten cycles of irradiation. However, it is recommended that this sequence of irradiations be terminated at the end of five cycles. Present plans are to start a new sequence of irradiations in a substantially harder flux with four fresh samples.

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1.0 Introduction

The program of thorium irradiation at the Materials Testing Reactor (MTR) has been described and its recent history has been reviewed elsewhere^(1,2). The present report describes some preliminary results on the irradiation of four samples containing about 60 grams of thorium plus varying amounts of U-233. Additional effort required to phase-out the program within a year is also described.

This is an interim report and will not include all calculational or analytical details since much of this work is not yet complete.

2.0 Description of Experiment

Irradiation and calibration samples were delivered during the summer and fall of 1965. Acceptance tests, such as ultrasonic testing for a metallurgic bond, and heat cycling for resistance to thermal stress were begun immediately. Pre-irradiation and calibration measurements in the Advanced Reactivity Measurement Facilities (ARMF I and ARMF II) were begun in October 1965, but were held to a minimum because of budgetary limitations.

Two pictorial views of the sample are shown in Figures 1 and 2. The first is a cutaway view showing sample construction, and the second shows the sample in its centering fixture ready for insertion into an X-basket.

2.1 Irradiation Program

Table I gives a description of the samples used in the irradiation experiments. They were chosen to have a spread of U-233 contents such

as to include the estimated value of the so-called equilibrium U-233 content. In the irradiation flux chosen this value was expected to be about 1.3% of the initial Th-232 content.

The first irradiations were begun during the latter part of November 1965 in a reflector location in the MTR. The MTR irradiation cycle consists of 16 or 17 days in the reactor and 4 or 5 days of down time for refueling and experiment change out. The irradiation schedule consisted of one MTR cycle of irradiation followed by one cycle out of the reactor for ARMF measurements.

Table I

Sample Composition

Sample Number	Nominal Content (U-233 wt %)	Thorium wt (gms)	U-233 wt (gms)
7	0.0	61.582	0.00
22	1.0	64.468	0.6487
29	1.3	63.748	0.8200
36	1.6	62.255	1.0016

A minimum of ten irradiation cycles is presently scheduled for these samples. Three such cycles have been completed as of the present writing.

The irradiating flux level was about 10^{14} n/cm²-sec. with a resonance to thermal flux ratio of about $1/23^{(7)}$.

The four samples were irradiated in pairs; two were undergoing irradiation during alternate MTR cycles while the other two were being measured in the ARMF I.

2.2 Measurement Program

Measurements are currently being made in three locations in ARMF I. None are being made in the ARMF II. Preliminary measurements in the ARMF II showed that that reactor was too sensitive to poison to permit the measurement of all samples with only one shim setting. The decision not to measure them in ARMF II was based on the fact that the necessity for changing the shim setting would compromise the accuracy, as well as increase the time required to get a measurement. Since a great many measurements would be needed, time became an important factor. The three locations used were the center of the ARMF I water hole (1), a side location (9), and a lattice location (6). These are shown in Figure 3.

Reactivity measurements were made on the unirradiated samples prior to irradiation and at least twice between irradiation periods. The first post-irradiation measurement was made four or five days after removal from the MTR, after the xenon transient had died out. The second measurement was made about two weeks later, or just prior to re-insertion into the MTR. Standard comparison samples were also measured at these times to detect any variation in ARMF I operating characteristics. Upon the completion of the scheduled irradiations, measurements will be made at about one month intervals for a total decay time of eight to ten months.

Calibration consists of an extensive series of measurements on the following types of samples:

Zirconium only (cladding material)

Thorium-standard (approximately same content as irradiation samples)

Thorium-calibration (contain various amounts of pure thorium)

Th-232 and U-233-calibration (same content as irradiation samples)

Th-232 and Boron-calibration

Th-232 and Gold-calibration

Mixtures-Th-232 and U-233 and Boron or Gold in varying amounts

2.3 Calculational Program

At the present writing the calculational program is incomplete. This section, therefore, will only outline the approach being used, in-so-far as the analysis of content of the irradiated samples is concerned. Details will be given of calculations dealing with the results reported here-in.

The calculational procedures are of two kinds and these will be issued as separate reports when they have been completed. One deals with the calibration and leads to a calculation of sample content from reactivity measurements. The other deals with a prediction of sample content from irradiation history and the nuclear constants of the produced isotopes. The latter problem has been discussed at length in the literature^(3,4,5).

The calibration work will deal with such effects as absorption and production of neutrons in three groups (thermal, intermediate, and fission) group-to-group scattering, water displacement, and neutron leakage. The various samples have been designed to have one or more of these effects dominant. By means of a least square fit to the derived equation, the reactor response factor of each of these effects can be evaluated. It is hoped that some of them might be negligible or compensatable by a suitable correction technique. The goal of these studies is to derive, if possible a procedure for calculating effective sample cross-sections for the three energy groups from reactivity measurements made in the three reactor locations. A comparison of

these cross sections with those calculated from the predicted content provides a means for testing cross-sections and calculational procedures.

Following irradiation the sample contains, principally, thorium, U-233, Pa-233, and in addition, small quantities of other irradiation products. After the xenon has had time to decay, any reactivity changes that remain are due principally to the decay of Pa-233 and the build-up of U-233. Both events produce an increase in reactivity with the transformation constant of Pa-233 since one U-233 atom is produced for each Pa-233 atom that decays. Hence, the measured reactivity transient can be described by the equation

$$\rho_t - \rho_o = (\rho_\infty - \rho_o)(1 - e^{-\lambda t}), \quad (1)$$

where

ρ_t = measured reactivity at any time, t ;

ρ_o = sample reactivity at termination of any irradiation period;

ρ_∞ = sample reactivity after a very long decay time (eight to ten months);

λ = decay constant for Pa-233

If measurements are made at two times, $t = t_1$ and $t = t_2$, then one has two equations from which either ρ_o or ρ_∞ may be eliminated yielding equations for ρ_o and ρ_∞ as follows:

$$\rho_o = \frac{\rho_{t1}[1 - \exp(-\lambda t_2)] - \rho_{t2}[1 - \exp(-\lambda t_1)]}{\exp(-\lambda t_1) - \exp(-\lambda t_2)} \quad (2)$$

$$\rho_\infty = \frac{\rho_{t2} \exp(-\lambda t_1) - \rho_{t1} \exp(-\lambda t_2)}{\exp(-\lambda t_1) - \exp(-\lambda t_2)} \quad (3)$$

The preliminary results quoted in this report are reduced to ρ_0 and ρ_∞ by the above equations. Some such procedure is necessary, because the time scale of the experiment requires that reactivity measurements be made during the Pa-233 decay period. Reduction to ρ_0 and ρ_∞ , therefore, provides a consistent basis for comparison of the sample content as a function of irradiation exposure.

3.0 Results

The results of reactivity measurements on the unirradiated samples ($nvt = 0$) and on the irradiated samples are shown in Figures 4 and 5. Except for the build-up of fission products and the burn-up of thorium, both of which are small during the early stages of irradiation, the U-233 content is nearly in direct proportion to the measured reactivity. In the absence, therefore, of a complete calibration the reactivity measurement can serve as a qualitative indication of the U-233 production. In Figure 4 the comparative reactivities, ρ_0 and ρ_∞ , as calculated from Eqs. (2) and (3), are shown as a function of accumulated exposure (nvt). Results for samples initially containing 1%, 1.3%, and 1.6% U-233 are shown in Figure 4; and for an initially 100% thorium sample the results are shown in Figure 5.

The difference between the ρ_0 and ρ_∞ curves represent the production of Pa-233. It appears that the Pa-233 content is approaching a constant value (equilibrium). Since the 1% curve has begun to rise, and the 1.3% curve is nearly constant it would appear that the "equilibrium" U-233 content will be close to 1.3%, which is the theoretical value for a thermal flux.

4.0 Discussion and Recommendations

The results quoted in the previous section are only qualitative and a reactivity versus fuel and poison content calibration will be required before quantitative results can be obtained. To derive meaningful results from these data a considerably more advanced calibration procedure will be needed than has previously been used in other ARMF programs. There are several reasons: (1) The infinite dilution technique is not applicable and, therefore, the effects of flux depression and shielding within the sample must be considered. (2) Thorium has a resonance integral in excess of ten times the thermal cross section and hence the single absorption group model is not applicable. (3) Other experiments in the ARMF have shown that scattering effects in massive metal samples may amount to several μk of reactivity. (4) Previous experiments have also shown that fast fission in thorium may also make a significant contribution to the observed effects. A three group analysis is, therefore, indicated, since three is probably the maximum number of groups that can be handled in the ARMF I reactor because of limitations on suitable measuring locations. More than three measuring locations are available in the ARMF I, but the reproducibility is not of equal quality in all of them. Furthermore, the increased cost of making more measurements and a reduced contrast in sensitivity among the various locations leads to a state of diminishing returns as the measuring locations are proliferated. A major effort will, therefore, be required during the coming year to reduce these data to quantitative results.

The calculation of irradiation products is one phase of the Th-232-U-233 problem that has received much attention in the literature. The calibration discussed in the previous paragraph leads directly to

a comparison between the experimentally measured and the predicted content of an irradiated sample. Excessively prolonged irradiations should not be necessary to establish this agreement between theory and experiment. In fact, even the qualitative results quoted in the previous section are pretty much what was expected; and it is questionable whether continued irradiation of these samples will yield information of value commensurate with the cost.

Previous calculations⁽⁶⁾ have shown that the thorium resonance integral plays a significant role in determining the so-called equilibrium U-233 content. It is expected, therefore, that irradiations in a harder flux than is presently being used might yield more useful information than might be expected from the current series. For these reasons plans for the irradiation of four samples in a harder flux are being formulated. U-233 contents of 1.3%, 1.6%, and 1.9% in addition to 100% thorium are contemplated. Five cycles of MTR irradiation ought to be adequate to provide the desired information. Since these samples will be alternated as before, five MTR cycles of irradiation per sample will be required for a total of thirty calendar weeks. This work could, therefore, be completed within the year remaining in accordance with present Atomic Energy Commission policy.

The calculational and calibration work discussed above can be carried on concurrently with the irradiations. A minimum time should, therefore, be needed to complete the reporting phase after completion of the irradiation phase.

Long term decay measurements to confirm the results predicted by Eqs. 2 and 3 can also be carried out concurrently with the hard flux irradiations.

Chemical analyses are recommended for selected samples on completion of all irradiations and subsequent reactivity measurements.

5.0 Conclusions

Since the present AEC policy is to phase out this program within the following fiscal year, a program to accomplish this has been outlined. The recommended program consists of the following:

1. Terminate current irradiations in the MTR at the completion of five irradiation cycles.
2. Continue for about ten months the reactivity measurements for tracing the decay of Pa-233.
3. Chemical analyses to confirm calculations.
4. Irradiate four samples in a hard flux for an accumulated nvt approximately equal to that in the softer flux.
5. Make concurrent calibration measurements and predictions of contents.

All experimental work outlined above can be completed by the end of fiscal 1967. A few months more should be allowed for the completion of chemical analysis, data reduction, and writing a final report.

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Figure 1 - Sample Geometry

PPCo-A-4082

Figure 2 - Sample with centering fixture.

Figure 3 - Reactor core showing measurement locations.

Figure 4 - Comparative Reactivities -
Initial content: 1.0% U^{233} , 1.3%
 U^{233} , 1.6% U^{233} in thorium.

Figure 5 - Reactivity changes 100%
thorium.

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