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FFTF COBALT TEST ASSEMBLY RESULTS

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Abstract: A cobalt test assembly containing yttrium hydride pins for neutron moderation was irradiated in the Fast Flux Test Facility during Cycle 9A for 137.7 equivalent full power days at a power level of 291 MW. The 36 test pins consisted of a batch of 32 pins containing cobalt metal to produce Co-60, and a set of 4 pins with europium oxide to produce Gd-153, a radioisotope used in detection of the bone disease Osteoporosis. Post-irradiation examination of the cobalt pins determined the Co-60 produced with an accuracy of about 5%. The measured Co-60 spatially distributed concentrations were within 20% of the calculated concentrations. The assembly average Co-60 measured activity was 4% less than the calculated value. The europium oxide pins were gamma scanned for the europium isotopes Eu-152 and Eu-154 to an absolute accuracy of about 10%. The measured europium radioisotope and Gd-153 concentrations were within 20% of calculated values. In conclusion, the hydride assembly performed well and is an excellent vehicle for many Fast Flux Test Facility isotope production applications. The results also demonstrate that the calculational methods developed by the Westinghouse Hanford Company are very accurate.

Introduction

The Fast Flux Test Facility (FFTF) is a sodium-cooled, mixed oxide fueled fast reactor operated for the U.S. Department of Energy (DOE) by the Westinghouse Hanford Company (Westinghouse Hanford) near Richland, Washington. The primary FFTF mission is to test fuels and materials in support of the U.S. Liquid Metal Reactor (LMR) program. The FFTF achieved full power operation at 400 MW in 1981 and has an excellent operational history. A gradual decline in emphasis on the U.S. LMR Program has led to an attempt to identify potential alternate missions for the FFTF. One such mission is the Isotope Production Program, in which novel methods for producing radioisotopes useful in medical, military, and industrial applications are being explored.

Radioisotopes identified early in the program include Co-60 and Gd-153. The radioisotope, Co-60, is valuable because of its energetic gamma rays and is used for sterilization of medical supplies. In addition, there are potentially large applications, such as food and sewage irradiation. The market for Co-60 is presently supplied primarily by the Atomic Energy of Canada Ltd. from the Canadian CANDU reactors. Thus DOE has expressed an interest in developing domestic supply alternatives. Initial estimates of Co-60 production in the inner row of radial reflectors in the FFTF indicated that only low quality Co-60 would be produced, in the neighborhood of 20 Ci/g. The desired quality is 100 Ci/g. Subsequent detailed Monte Carlo physics calculations showed that production could be enhanced by approximately a factor of five by introducing hydrogen into the assembly to provide neutron moderation of the fast neutron FFTF spectrum, thereby achieving an epithermal neutron spectrum. Because most isotope production reactions of interest to reactors involve neutron absorption events, which typically have large cross section resonances in the energy range below 1 keV, it is obvious that in most cases a hydride assembly is superior.

After a survey of methods to introduce hydrogen into an FFTF assembly, Westinghouse Hanford decided to use pins containing yttrium hydride pellets, interspersed with the target pins. It was immediately evident that the hydride assembly had the potential of causing power peaking in neighboring fuel assemblies because of return low energy neutrons, which have large fission cross sections. A series of experiments at Argonne National Laboratory (Idaho Falls, Idaho) in the Zero Power Plutonium Reactor (ZPPR) validated the methods used by Westinghouse Hanford to calculate power peaking.[1] The experiments were successful and proved the detailed physics calculations are usually accurate to better than 10%. This was to be the last major hurdle to irradiation in FFTF.

The cobalt production physics calculations also showed that high quality Gd-153 could be produced in one cycle of irradiation, and four pins containing europium oxide pellets were included in the assembly. The High Flux Isotope Reactor (HFIR) at Oak Ridge National Laboratory (ORNL) had produced Gd-153 previously, but it was shut down in 1987 because of safety concerns. A dual photon source, Gd-153, for detection of the bone-thinning disease, Osteoporosis, is produced by a two-step capture process: Eu-151 captures a neutron to produce Eu-152 in a short-lived metastable state, which decays to stable Gd-152, which captures another neutron to form Gd-153. The product quality is expressed as Curies of Gd-153 per gram of Gadolinium (Ci/g). Typical acceptable quality factors for ORNL HFIR irradiations are in the range 50 Ci/g, and initial estimates indicated the potential for better quality Gd-153 in the FFTF hydride assembly.

The test assembly was irradiated for one cycle with the following purposes:

- Demonstrate the accuracy of the physics calculations
- Produce Gd-153 for medical applications
- Demonstrate the ability to handle high activity Co-60 pins at the FFTF
- Provide yttrium hydride pins for post irradiation examination to determine the stability of yttrium hydride under prototypic operating conditions.

Summary

Thirty-two cobalt pins and four europium oxide pins were irradiated in the Cobalt Test Assembly for 137.7 Equivalent Full Power Days (EFPD) in the FFTF during Cycle 9A at a power level of 291 MW. Following a decay period of about five months, the test pins were recovered in the FFTF Interim Examination and Maintenance (IEM) Cell and shipped to the Pacific Northwest Laboratory (PNL) Shielded Materials Facility (SMF) for reprocessing. The europium oxide pins were gamma scanned absolutely for Eu-152 and Eu-154 by comparison with a europium oxide standard. The cobalt pins were gamma scanned for Co-60. In addition, one cobalt pin was disassembled to recover thin cobalt wafers, which were dissolved and assayed for absolute Co-60 activity by PNL Radiochemistry. The europium oxide pins were packaged and shipped to ORNL for recovery, assay of the Gd-153, and sale by the ORNL Isotope Distribution Office.

Yttrium hydride pins were included in the Cobalt Test Assembly to moderate high-energy FFTF neutrons. While this increased the isotope production reaction rates, the mean-free path of the important low energy neutrons was much smaller than the physical mesh size normally used to analyze FFTF experiments. In addition, the neutron capture rates involved were very sensitive to the resonance energy range (below 1 keV).

This sensitivity mandates the use of sophisticated calculational techniques to predict final isotope activities. The technique developed at Westinghouse Hanford for this type of problem uses the Monte Carlo code MCNP, a continuous energy neutron/gamma ray transport code, with the capability to model the Cobalt Test Assembly geometry exactly. The isotopic capture rates were calculated using ENDF/B-V cross section data for cobalt and europium. The gadolinium cross sections were modified based on recent data. Predictions of cobalt, europium, and gadolinium activation were performed to a statistical accuracy of better than 10% for comparison with measurements.

Calculational Methods and Results

The calculational method used to predict reaction rates in the cobalt test assembly was developed at Westinghouse Hanford and is described in reference 1. The pointwise (in energy) Monte Carlo code MCNP[2] first calculates boundary currents on a cell containing the target assembly and six surrounding assemblies, using a model including the entire FFTF reactor as loaded for Cycle 9A. For this initial calculation, the target assembly was homogenized. This whole-core calculation typically requires several hours of CRAY computer time to obtain adequate statistics. The currents from the first MCNP run drive a cell calculation, in which the target assembly is modeled in detail. The surrounding six assemblies were modeled to the extent desired, depending on information required. The modeling of the cobalt test assembly was almost exact, with each pin modeled separately; even the cladding was modeled explicitly.

Reactor material cross sections used in all MCNP calculations were from ENDF/B-V. The europium, gadolinium, yttrium, cobalt, and hydrogen cross sections used in the reactor calculations were from ENDF/B-V, except for the epi-thermal cross section for Gd-152, which was modified based on information from reference 3. The reaction rates computed in the MCNP cell calculations for mid-cycle 9A conditions were used to calculate end-of-cycle amounts and activities of all relevant isotopes. There were two adjustments to reaction rates from MCNP.

1. The fractions of Eu-151 capture events leading to the ground and metastable states of Eu-152 are 0.65 and 0.35, respectively.

2. An approximate adjustment in the epi-thermal cross section for Gd-152 capture based on information from reference 3 was also made after the MCNP calculation.

Europium Oxide Pin Measurements

The europium oxide pins were all gamma scanned in the 324 building SMF facility. A germanium gamma-ray detector in anti-coincidence with a Compton suppression annular sodium-iodide detector measured the gamma ray spectrum using a multichannel analyzer. The gamma ray lines listed in Table 1 were analyzed for Eu-152 and Eu-154 activity by comparison with a standard one-inch long europium oxide source pin, which has an activity of about 1 Curie each of Eu-152 and Eu-154. The detector viewed the pin through a collimated slit and shield installed in a hot cell window. The slit width used for these measurements (test pins as well as source pin) was 0.254 mm. The absolute calibration uncertainty is about 10% (one sigma), and the overall reproducibility of the counting system is better than 5%. The pins were automatically moved in 5.08 mm axial steps between counts. Gamma ray peak area data were recorded on magnetic tape and hard disk, and transferred to floppy disks for analysis and plotting. The counting time at each location was 120s.

Table 1. Europium Gamma Ray Analysis Lines

EU-152 LINES (keV)	EU-154 LINES (keV)
779	723
964	757
1086	996
1112	1005
1408	1275

Of all the isotopes produced, only gamma rays from Eu-152 and Eu-154 were resolvable. For each isotope, five gamma ray lines were used in an unbiased average to calculate the activity per unit length, with units Curies/cm. Plots of the analyzed data for pin E4 for Eu-152, compared with the calculational methods described in the previous section are shown in Figure 1. Pin E4 was on an assembly flat adjacent to row-6 fuel. The comparison between data and predictions for the other europium oxide pins shows similar agreement. In general, predictions and measurements agree within 20% with a general tendency to overpredict activity in the central pin section.

Following gamma scanning of the europium oxide pins, each pin assembly was sheared at the necked regions to form three sections each. The pin sections were packaged and shipped to ORNL for recovery of the gadolinium. The gadolinium was chemically separated from the europium oxide, and

the resulting gadolinium was assayed for Gd-153 content by absolute gamma ray counting. Figure 2 illustrates predicted and measured Gd-153 quality for pin E3 at the end of Cycle 9A, as well as on September 1, 1987. The ORNL experimental data in Figure 2 are represented on vertical lines in the middle of the upper two pin sections because it is not known, within a section, where the analyzed pellets were located. The spread of

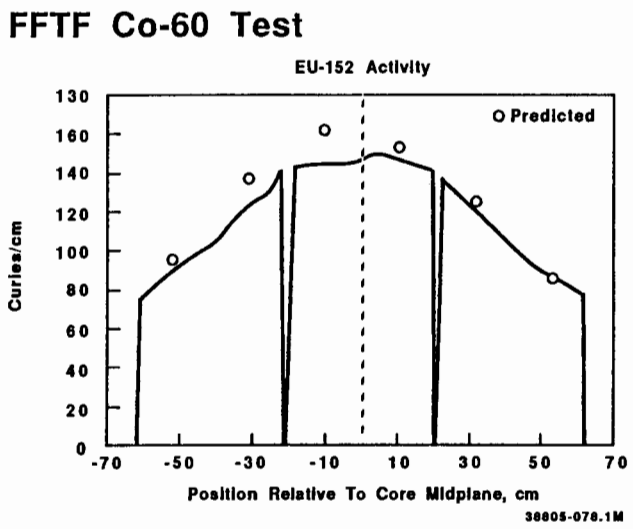

Figure 1 Eu-152 activity for Pin E-4

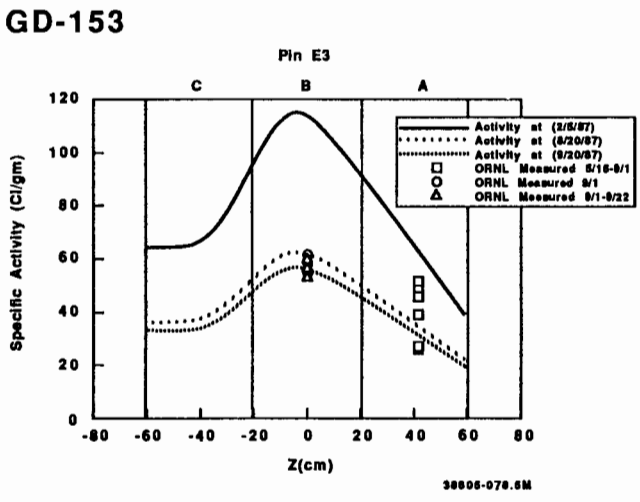

Figure 2 Gd-153 quality for Pin E-3

measured values is in good agreement with the predicted quality range for the middle and upper sections.

Cobalt Pin Measurements

The cobalt pins were gamma scanned in exactly the same manner as that described for the europium oxide pins, with the exception that only two gamma ray lines (1,173 keV and 1,333 keV) were analyzed for Co-60, and each count interval was 60s. An additional absolute calibration point was obtained by counting two thin cobalt wafers (from the center pin section of pin 32) for gamma rays on a calibrated counting system in the PNL Radiochemistry laboratory. The calibration obtained from the cobalt wafers agreed within 10% with that based on the europium oxide standard source. The measured and predicted data for pin 32, which was near the middle of the test assembly is presently in Figure 3. Measured and predicted Co-60 concentrations agree within 20%. The calculated assembly average Co-60 concentration is 16.65 Ci/g, and the measured value is 16.01 Ci/g. The calculated axial peak to average Co-60 concentration is 1.44, and the measured value averaged over all 32 pins is 1.37 (with a measured range of 1.31 to 1.43). There was no apparent systematic variation of the measured axial peak to average value among various pin groupings.

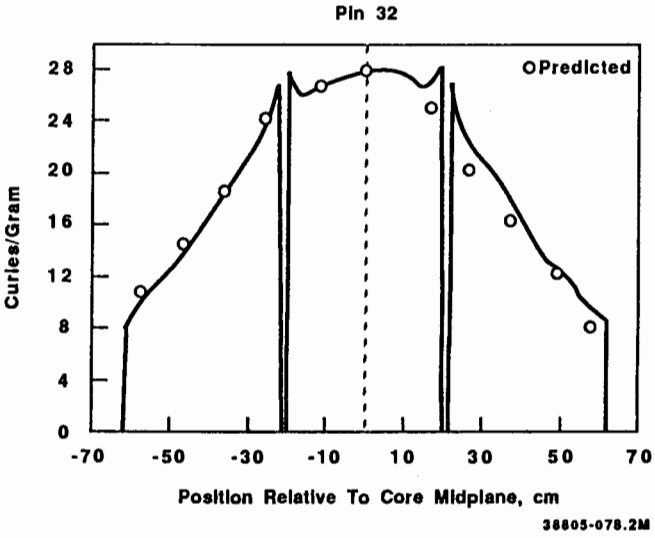

Figure 3 Co-60 Activity for Pin 32

Conclusions

Reaction rates predicted in the FFTF hydride cobalt test assembly with the MCNP calculational methods developed by Westinghouse Hanford are within 20% of experimental measurements. This difference is comparable with the combined nuclear cross section, measurement calibration, and MCNP statistical uncertainties. In addition, the test showed that production pins in a hydride assembly in a high flux fast reactor can make marketable amounts of high quality radioisotope material. The cobalt test assembly was in a relatively low flux row 7 radial reflector location. Relocation to an inner core position (with two to four times the total flux) would result in producing more and higher quality material. Such relocation would raise several issues:

- Impact on other programs
- Larger reactivity penalty
- Higher neutron charges

- Performance of the yttrium hydride moderator pins under more extreme conditions

- Design changes to mitigate overheating in neighboring fuel pins.

In particular, production of Co-60 in row 7 of FFTF of Co-60 with a quality in the range 70 to 100 Ci/g is achievable with long enough irradiation times (five to six cycles). Irradiation of europium oxide pins for two reactor cycles in row 7 would yield large amounts of Gd-153 with a quality at shutdown greater than 200 Ci/g, which is much higher than any thermal reactor can produce.

The next phase of developing the FFTF Isotope Production mission includes identification of other useful and marketable isotopes that can be efficiently produced in FFTF[4], followed by design and fabrication of a modified hydride production assembly and irradiation of a variety of targets.

References

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