HEDL-SA-2557-FP

Conf-820406--21

CONF-820406--21

DE82 017527

MASTER

DESIGN OF UNIQUE PINS FOR IRRADIATION OF HIGHER ACTINIDES

J. A. Basmajian, K. R. Birney, E. T. Weber, HEDL
H. L. Adair, T. C. Quinby, S. Raman, ORNL
B. C. Bateman, J. K. Butler, K. M. Swanson, UKAEA

March, 1982

ANS Topical Conference on Fast, Thermal, and Fusion Reactor Experiments

NOTICE AMERICAN NUCLEAR SOCIETY

April 12-15, 1982
Salt Lake City, Utah

HANFORD ENGINEERING DEVELOPMENT LABORATORY
Operated by Westinghouse Hanford Company, a subsidiary of Westinghouse Electric Corporation, under the Department of Energy Contract No. DE-AC06-76FF02170
P.O. Box 1970, Richland, Washington 99352

MN ONLY

Research at ORNL sponsored by USDOE-RRT under Contract No. W-7405-eng-26 with the Union Carbide Corporation

DESIGN OF UNIQUE PINS FOR IRRADIATION OF HIGHER ACTINIDES IN A FAST REACTOR

J. A. Basmajian - WHC/HEDL
K. R. Birney - WHC/HEDL
E. T. Weber - WHC/HEDL
H. L. Adair - ORNL
T. C. Quinby - ORNL
S. Raman - ORNL
J. K. Butler - UKAEA
B. C. Bateman - UKAEA
K. M. Swanson - UKAEA

ABSTRACT

The actinides produced by transmutation reactions in nuclear reactor fuels are a significant factor in nuclear fuel burnup, transportation and reprocessing. Irradiation testing is a primary source of data of this type. A segmented pin design was developed which provides for incorporation of multiple specimens of actinide oxides for irradiation in the UK's Prototype Fast Reactor (PFR) at Dounreay Scotland. Results from irradiation of these pins will extend the basic neutronic and material irradiation behavior data for key actinide isotopes.

I. INTRODUCTION

The actinides produced by transmutation reactions (Figure 1) in nuclear reactor fuels are a significant factor in nuclear fuel burnup, transportation and reprocessing.[1] Measurements of actinide fission and capture reaction rates in fast reactor spectra are needed to provide improved data for fast reactor analysis and for evaluation of postirradiation processes sensitive to formation and depletion of actinide isotopes. Reactor irradiation of higher actinides has been limited, both in extent and quantities of material irradiated. Nuclear cross section data, yield data and methods of burnup by fissioning or conversion to short-lived isotopes are of interest. Based on mutual recognition of the paucity of irradiation data in this area, a joint US/UK irradiation program was established to extend the basic neutronic and irradiation behavior data on key actinide isotopes.

A segmented pin design was developed which provides for incorporation of multiple specimens of a number of actinide isotopes for irradiation in the UK's Prototype Fast Reactor (PFR) at Dounreay. Since this test was the first to incorporate more than milligram quantities of actinide isotopes in an irradiation experiment, a number of design, fabrication and handling problems were addressed in the course of test preparation. These activities were undertaken as a joint effort by Westinghouse Hanford Company (WHC) and Oak Ridge National Laboratory (ORNL) in the US working with staff of the United Kindgom Atomic Energy Authority (UKAEA) at Dounreay and Risley (Figure 2).

II. DESIGN CONSIDERATIONS AND FEATURES

The objective of the irradiation program is to verify the neutronic and irradiation performance of americium and curium oxide in a fast reactor. The data obtained from the test will be used: 1) to define waste management options and basic neutronic data, and 2) to assess the irradiation potential of the oxides as fuel materials. Two types of test samples are involved. One type comprises milligram quantities of high purity, single isotopes of the higher actinides designed to provide basic physics data for the fast flux environment. The second type of sample consists of fissionable actinide isotopes in gram quantities and in fuel pin configuration, to explore behavior related to reirradiation or recycling of higher actinide isotopes in nuclear fuel. In addition, neutron dosimetry capsules were recognized as desirable components of the irradiation to characterize the specific neutron flux at the test position. The major focus of the design effort was to establish a test pin configuration and related materials specifications suitable for an irradiation covering several PFR operating runs.

A. TEST DESCRIPTION

The test was defined to consist of four pins, with a configuration compatible with a standard PFR Demountable Subassembly (DMSA) irradiation vehicle (Reference 2). These special pins replace one or several regular PFR fuel pins in the standard 19 pin, grid spaced cluster. The component sections for makeup of the pins were specified as follows:

1) A fuel section, containing gram quantities of cylindrical pellets of actinide oxides with a single cladding and gas plenum space typical of fuel pins.

2) A physics specimen section, containing multiple small capsules, each of which is loaded with milligram quantities of single isotope actinide oxides.

3) A dosimeter section, containing flux wires and spectral sets prepared and analyzed consistent with well established methods (Reference 3).

Two identical pins were designed to provide for irradiation of the specimens to different exposure (burnup) levels. These two pins were made to contain three actinide pellet sections, four dosimetry sections and one physics section. The three actinide pellet sections contain these pellet types: 1) curium oxide, 2) americium oxide and 3) a mixed Cm, Am plus lanthanide oxide. The third pin consists of a set of the three actinide oxide pellet sections plus dosimeters, with no physics section; the fourth pin has only a physics section, with a dosimeter section at either end. These last two pins are intended to provide flexibility for achieving a different exposure level for one set of pellet and physics specimens, such as an extended irradiation for the physics specimens to greater than the 180 full power day (FPD) goal exposure for the test.

Nondestructive examination, followed by destructive examination of pellet sections are planned to be performed in the UK. The pins will be cut to separate the dosimetry and physics sections and permit shipment of these pin sections to the US for analyses. Specific features of the pin design were developed consistent with logistical considerations. Materials and component preparation, pin segment fabrication and partial assembly of pins in the US were performed at both ORNL and WHC. Size of the available shipping cask precluded shipment of full length pins so partially assembled pin segments were shipped to the UK for final assembly and irradiation.

B. SPECIFIC DESIGN FEATURES

The general pin configuration (Figure 3 and 4) shows a screw connection and reduced diameter region between each cladding section. The reduced diameter was necessary to maintain the configuration to permit use of a standard end-cap to cladding weld. This screw connection was designed to allow for fabrication of each pin segment as a separate entity, and then be connected with other segments to make up the full length pin. This connection represented a key element of the segmented pin design since: 1) it provided the capability for reconstitution of pins, 2) it permitted loading of individual pin segments one at a time, avoiding or reducing the shielding problems related to the handling of full length pins, and 3) it allowed shipment of segments about 50 cm in length to the UK rather than full length pins. All actinide oxide and physics samples were located axially to be within the reactor core and the axial arrangement of the actinide oxide sections was made the same in all pins.

One problem related to the reduced diameter region involves the grid dimples in the DMSA. The design criteria required assurance that the full diameter cladding always be in contact with the grid dimples during irradiation. Allowances for axial movement of the pins in the DMSA and irradiation induced growth of the pins were included in the design analysis. This requirement was compounded by the need to make all the actinide sections the same length for ease of fabrication, include dosimeter sections at strategic locations, and place the physics section within the axial bounds of the fueled region. The design of the pin satisfied the requirements while accommodating about 35 mm of potential axial motion due to "pin flotation", in addition to thermal and irradiation induced growth effects.

Cladding used to make all the sections was Ti modified 316 SS, which is expected to show little irradiation induced growth after two runs in PFR.[4] The minimum cladding thickness is 0.76 mm. This cladding thickness results from the use of a funnel for loading the pellets to avoid contamination of the closure weld at one end of each section. At the location of the 2.54 cm pellet stack, the wall thickness is 0.94 mm. The actinide sections had the same axial correspondence in each of the three pins and are located in the upper half of the PFR core region. Both the physics and dosimeter specimens were enclosed in their own miniature subcapsules which were sealed inside cladding of the same type as that used for the actinide oxide specimens. The separate sections of each pin are of different lengths. The actinide oxide sections are about 13 cm long, the dosimeter sections are about 8 cm long and the physics sections are about 35 cm long (for Pin 1 and Pin 2) or

about 45 cm long (for Pin 4). Each pin section is assigned a unique identification number engraved on the end cap. There are also top and bottom pin extensions to achieve an overall pin length consistent with the fuel pins making up the remainder of the clusters.

The minimum cladding thickness requirements established one of the bounds influencing the actinide oxide pellet dimensions and specifications. Additional criteria were applied which represented an interest in establishing thermal conditions in the pellets which would provide a basis for a comparison of actinide pellet irradiation behavior to the mixed U, Pu oxide fuel performance data base.[5] The criteria imposed were: 1) cladding for the actinide oxide sections will operate near 550°C and 2) actinide oxide pellet centerline temperatures will be below 2200°C and above 1000°C.

III. MATERIALS AND FABRICATION

All actinide materials for the test were supplied and prepared by ORNL.[6] Specifications for the pellet specimens were developed by WHC based on analogy to specifications used for mixed UO_2-PuO_2 fuel. The sesquioxides $^{241}Am_2O_3$, $^{244}Cm_2O_3$ were specified to contain >85% of the isotope of primary interest. A third sesquioxide was specified with metal components $^{241}Am_6$ $^{244}Cm\ \bar{La}_7$ where $\bar{La}$ corresponds to a mixture of stable lanthanide elements of La, Ce, Nd, and Sm in the mass ratio 1:2:3:1. Control of stoichiometry for an oxygen to total metal ratio of 1.50 ± 0.02 was specified to provide a stable oxide phase and a uniform basis for design and analysis. Impurity

specifications were generally consistent with those for nuclear fuels, but because of the decay products for these isotopes, most lanthanides and all other actinides were not considered as impurities. Hot pressing in graphite dies was the method employed by ORNL to fabricate right circular cylinder pellets with bulk density of 90 ± 5% TD. Pellets were hot pressed to the specified length and diameter, and maintained carbon impurity content well within a specified carbon + sulfur limit of 500 ppm. Microstructure features and a thermal stability test at 1600°C were specified with the intent to minimize the potential for significant in-reactor pellet densification during irradiation. Although the high radiation levels associated with ^{241}Am and ^{244}Cm required special handling procedures, post-fabrication pellet characterization results obtained at ORNL and WHC showed remarkable consistency with specifications. For such a unique fabrication project the results were considered exceptional.

Specimens of all twenty-one actinide isotopes to be evaluated for physics data were prepared and loaded as oxide powders. Specified milligram quantities of these isotopes were loaded into thin wall (0.5 mm) vanadium capsules using precise weighing techniques. Each vanadium capsule was sealed by a fusion weld, leak checked and uniquely identified by a dot code impressed on the capsule. This method has been used successfully by ORNL in prior tests.

Fabrication of the actinide specimens and closure of the pin sections was completed at ORNL as a joint effort between ORNL and WHC. Assembly of the

pin sections into ~50 cm long segments was completed at WHC and witnessed by personnel from ORNL and UKAEA. Assembly of these components required use of shielded equipment as the actinide isotopes have significant gamma and spontaneous fission neutron activity. Dose rates for the unshielded Cm_2O_3 pin section was ~600 rem/hr from neutrons at 1 cm. Use of the shielded welding box reduced the dose rate at the surface of the box to ~500 mrem/hr. At the normal working position the dose rate was <100 mrem/hr. Most of the exposure was encountered during insertion and removal of the pins from shielded containers. At 1 ft, the dose rate was 650 mrem/hr with no shielding. In general, very low accumulated dose was assessed by personnel in the US and the UK during pin assembly operations.

Due to the high activity associated with the Cm_2O_3 pellets, they were thermally hot and required special handling. This requirement related to control of contact interfaces as the cladding surface temperature was near 200°C after pin closure. Thermal aspects prior to irradiation are detailed in Section IV.

Am_2O_3 pellets have a lower heat generation rate because of the alpha decay ($T_{1/2}$ = 433a) and spontaneous fission ($T_{1/2} = 10^{14}$a) and were easily handled in terms of thermal considerations. However, there was considerable gamma activity involved.

The small amount of ^{244}Cm in the M_2O_3 pellets (7% of metal atoms) resulted in relatively benign thermal effects and the primary handling problem related to the gamma emitted by the ^{241}Am.

IV. DESIGN AND PERFORMANCE ANALYSIS

A. ACTINIDE OXIDE SECTIONS

1. Thermal Analysis-Prior to Irradiation

The Cm_2O_3 pellets produce significant heat due to alpha decay ($T_{1/2}$ = 18.1a) and spontaneous fission ($T_{1/2}$ = 1.25 x 10^7a). The calculated heat generation for hot-pressed pellets with 90% of theoretical density was 27 W/cm^3. Calculations of the temperature for a 3.81 mm diameter by 5.08 mm long pellet were made using the HEATING-5[7] code. The pellet was assumed to be seated on a stainless steel platen of 25.4 mm diameter by 3.81 mm thick and the calculated temperature was 85°C at the pellet-platen interface and 267°C at the top. Pellet temperatures near 200°C were observed when placed in cladding tubes.

2. Thermal Analysis - During Irradiation

Burnup of ^{241}Am and ^{244}Cm during irradiation results in a decrease in heat production from these isotopes as the irradiation progresses. As shown in Figure 1, ^{241}Am produces ^{242}Am, ^{242}Cm and ^{238}Pu during irradiation. These isotopes add to the overall heating such that the heat generation increases with time. Also, ^{244}Cm produces ^{245}Cm and exhibits a similar increase in heat generation. Details of the magnitude of the predicted increases are given below. It is interesting to note that mixed U, Pu oxide fuel in FFTF

produces ~175 w/g and the materials used in the actinide oxide pellets of the test pins in PFR produce from 95 w/g to 240 w/g.

Heat generation during irradiation in the PFR was calculated based on flux data provided by the UKAEA and ENDF/B-V cross sections.[8] The total predicted heating of $^{241}Am_2O_3$ and its reaction products increased over 180 FPD from 136 W/g to 175 W/g (Figure 5). The initial heating rate of 179 W/g for $^{244}Cm_2O_3$ and its reaction products increased to 240 W/g (Figure 6). The total heating for the M_2O_3 started at 95 W/g and increased to 123 W/g. Heat transfer calculations were made for the "as-built" geometry and material loadings for each of the three types of actinide oxide sections. Similar centerline temperatures were desired for the different sections. Since the Cm_2O_3 pellets generated the most heat, they were placed in the lower flux portion of the upper half of the core region, the M_2O_3 pellets were placed in the higher flux region and the Am_2O_3 pellets were placed axially between the others. This placement provided a more uniform temperature distribution than could be obtained otherwise.

The irradiation behavior of the actinide oxides is unknown, and this is one reason for the test. Assumptions were made by analogy to U, Pu mixed-oxide fuels concerning pellet restructuring and gap closure, and fuel pin thermal analysis models[9] used for mixed uranium-plutonium oxide fuels were applied with appropriate adjustments in properties. A value for heat-generation for each particular actinide oxide that was 25% greater than the calculated value was used to approximate worst case operating conditions with overpower.

On this basis, the maximum temperature calculated for the Cm_2O_3 pellets was 1705°C after 60 days irradiation even though the heat generation rate had not reached its maximum (Table 1). Without the restructuring assumptions relating to gap closure and center void formation, the temperature would increase with the heating rate and reach a maximum at goal exposure. The combination of heat fluxes, gap size, center void and heat rate change as a function of time resulted in a peak temperature for the Cm_2O_3 at 60 days. However, for the Am_2O_3 the temperature peak was 1927°C at 180 days and for the M_2O_3 it was 1595°C at 60 days.

3. Pressures

Pressure calculations were made for the "worst case" actinide (Cm_2O_3) pin for a 270-day reactor residence time. Helium generation from alpha decay was considered after encapsulation for 600 days and irradiation for 270 days, assuming a fission gas yield of 30% with 100% release. The calculated pressure was 13 MPa, giving a hoop stress well below 35 MPa. Hence, cladding stress was not a significant problem for this pin design.

4. Cladding Chemical Interaction in Actinide Pins

An assessment was made of the potential for chemical attack of the cladding in the heat generating actinide pellet segments. Since more thermodynamic data were available on americium oxide, it was employed as a model for all three pellet materials. The oxygen potenital for Am_2O_3, ~-50 kcal/mol O_2 at cladding operating temperatures, exceeds the oxidation threshold for the

TABLE 1

PELLET PEAK CALCULATED TEMPERATURES (°C)

Pellet	Irradiation Time 1 Day	5 Days	60 Days	180 Days
Cm_2O_3				
Centerline	1695	–	–	–
Center Void	–	1639	1705	1558
Restructured	–	1276	1346	1189
Am_2O_3				
Centerline	1778	–	–	–
Center Void	–	1637	1900	1927
Restructured	–	1255	1470	1409
M_2O_3				
Centerline	1421	–	–	–
Center Void	–	1438	1595	1532
Restructured	–	1138	1247	1146

various components of stainless steel. Thus, Am_2O_3 could cause cladding oxidation at the start of irradiation. With very low concentrations of Cs and Te available early in the irradiation, cladding attack should be uniform and its depth limited.[10]

The oxygen potentials for the Am_2O_3 and Cm_2O_3 are more than sufficient to oxidize fission-product cesium to Cs_2O, which can cause intergranular corrosion of stainless steel.[11] However, the rate of formation of cesium and hence the availability of Cs_2O will be low. As Cs_2O is formed or migrates to the pellet cladding interface, it should combine as ternary oxides with the cladding or fission product elements, thus limiting its participation in intergranular corrosion. For these conditions, cladding oxidation should continue in a uniform manner, by a diffusion process, and the depth of attack would be limited.

B. PHYSICS AND DOSIMETRY SPECIMENS

There are a total of forty-four (44) physics and dosimeter specimens in each of the three physics sections. All pins contain the same materials with only minor weight differences (Table 2).

1. Temperature Calculations

The most fissile isotopes were ^{233}U, ^{235}U, ^{239}Pu, ^{241}Pu and ^{243}Cm. ^{243}Cm has the highest fission rate. Assuming all the heat generating material was centered as a solid cylinder in the vanadium capsule with a helium

TABLE 2

PHYSICS SPECIMENS

Isotopes	Weight (mg)	Calculated Temperature Range (°C)
^{230}Th, ^{231}Pa, ^{234}U, ^{238}Pu, ^{241}Pu ^{242}Pu, ^{243}Cm, ^{244}Pu, ^{248}Cm	1–5	590 to 845
^{233}U, ^{235}U, ^{236}U, ^{241}Am ^{243}Am, ^{244}Cm, ^{246}Cm	5–10	650 to 870
^{237}Np, ^{238}U, ^{240}Pu, ^{241}Am	10–15	675 to 705
^{232}Th	18	649

atmosphere, the temperature at the cylinder center was 1399°C. This condition was unrealistic, since the material would not be expected to assume that configuration and would be in contact with the wall. Assuming a conductivity which is 90% of the fully dense actinide oxide, and assuming the material is a powder on the inner circumference of the capsule, results in significantly lower calculated temperatures (870°C). These two cases represent the extremes for thermal predictions.

2. Compatibility

Numerous irradiation experiments performed by WHC, in the EBR-II reactor, have used vanadium dosimetry capsules containing oxides of U, Pu, Np as the material to be activated.[3] These actinide oxides performed satisfactorily where capsule-actinide oxide interface temperatures were below 900°C.

3. Pressure Calculations

Pressure buildup in the physics capsules was of considerable import. Calculations indicate that the greatest burnup would occur in the ^{239}Pu specimen, where 23% of the material would be fissioned in 180 FPD. The calculated pressure was 4.2 MPa after 180 FPD of fission. Using simple thin-wall stress calculations, this pressure translates to a stress of 8.42 MPa. For 270 days operation, Pin 4 would have 35% of the ^{239}Pu fissioned and the capsule pressure would be 4.35 MPa with a wall stress of 8.93 MPa.

Vanadium retains its strength unusually well at elevated temperatures. Although there were sparse high-temperature data, Reference 12 indicated a yield strength of ~137.8 MPa at 600°C. Vanadium encapsulated dosimeters used in EBR-II have operated in the 6.89 MPa range satisfactorily, and were selected for this application.

4. Dosimetry

The ORNL dosimetry was incorporated into the Physics Specimens section of the pin. The amounts and types of materials being used in these dosimeters were such that their operating conditions were much less severe than those in the Physics Specimens.

The HEDL dosimetry was incorporated into three identical dosimetry pin sections. Sophisticated thermal analysis of this configuration was not completed. Because of the small amounts of materials involved in the heating, a thermal calculation made by simplifying the geometry was acceptable, giving a temperature ~100°C above the coolant operating temperature.

V. CONCLUSIONS

The segmented pin design was developed to accommodate a variety of small test samples. It permitted fabrication of individual segments and avoided shielding and handling problems related to the full-length pins prior to final assembly.

The final pin configuration contained all the required specimens and located them in the PFR core such that similar temperatures of the actinide oxides were achieved and the grid dimple interface imposed by the DMSA was satisfied. This pin configuration would be readily adaptable to other experiments.

Fabrication techniques were established and verified. Assembly techniques required special shielding, but were accomplished with relatively low dose to all personnel.

VI. REFERENCES

1. "W. Hage First Technical Meeting on the Nuclear Transmutation of Actinides," EUR-5897, 1978.

2. Public Relations Branch, UKAEA Reactor Group, "Dounreay PFR Irradiation Service," February 1974 (Printed by Hutton, Hartley and Co. Ltd. Manchester).

3. E. P. Lippincott and J. A. Ulseth, "High Flux-Fluence Measurements in Fast Reactors," Proceedings of the Second ASTM-Euratom Symposium on Reactor Dosimetry, NUREG/CP-0004, p. 271, (1978).

4. C. M. Cox, R. J. Jackson and R. J. Neuhold, "Development of High Burnup LMFBR Fuels," HEDL-SA-2525-FP, Published in Nuclear Engineering International, November 1981.

5. International Conference on Fast Breeder Reactor Fuel Performance, Proceedings of the ANS Topical Meeting, Monterey CA, March 1979 ISBN: 0-89448-105-3.

6. T. C. Quinby, et al., "Preparation of Actinide Specimens for the US-UK Joint Experiment in the Dounreay Prototype Fast Reactor," ORNL-5858.

7. W. D. Turner, et al., "HEATING-5," ORNL/EDS/TM-15, March 1977.

8. F. M. Mann and R. E. Schenter, "HEDL Evaluation of Actinide Cross Sections for ENDF/B-V," HEDL-TME 77-54 (1977).

VI. REFERENCES (Cont'd)

9. D. S. Dutt and R. B. Baker, SIEX - A Correlated Code for the Prediction of Liquid Metal Fast Breeder Reactor (LMFBR) Fuel Thermal Performance, HEDL-TME 74-55, June 1975.

10. M. G. Adamson, "Mechanisms of Fuel Cladding Interaction: US Interpretation," Paper Presented at IAEA Specialists Meeting on Fuel Cladding Interaction, Tokoyo, Japan, February 21-25, 1977.

11. Journal of Nuclear Materials, 44, 96 (1972).

12. Aerospace Structural Materials Handbook, Vol 5, (1974).

NEUTRON IRRADIATION OF ^{244}Cm AND ^{241}Am

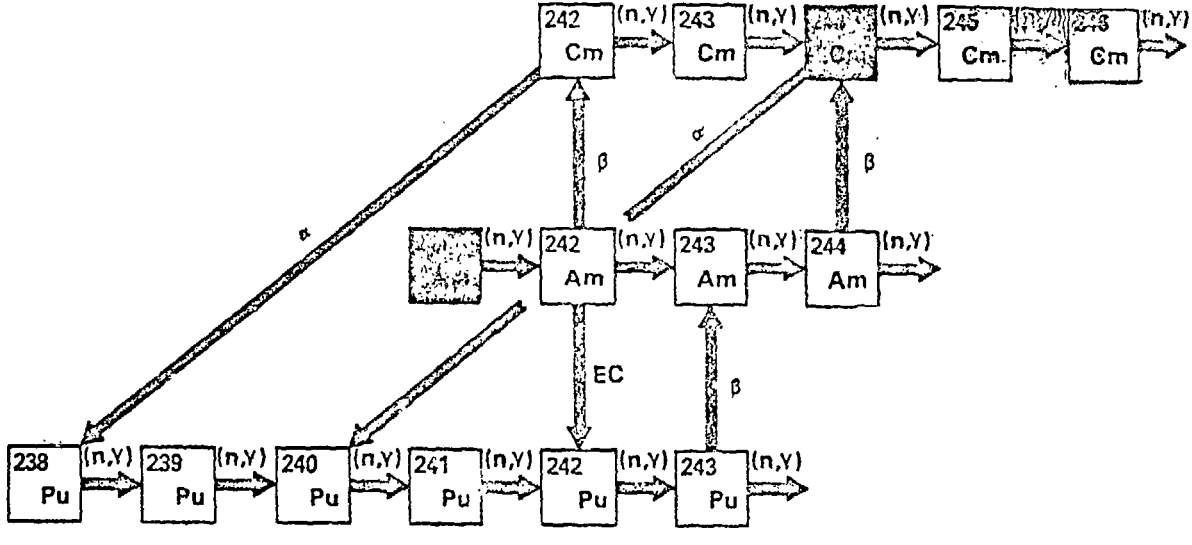


FIGURE 1. Neutron Irradiation of ^{244}Cm and ^{241}Am. Neg 8003293-1

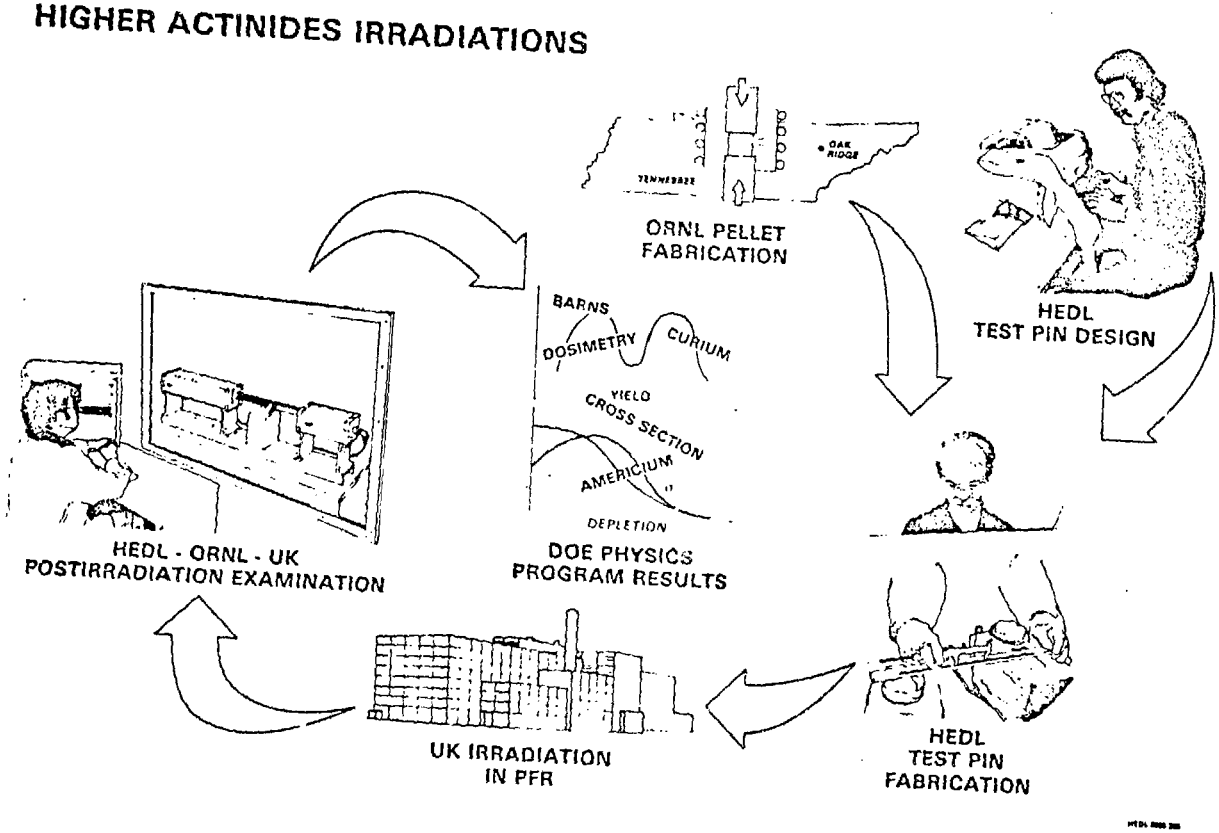


FIGURE 2. Higher Actinides Irradiations. Neg 8007830-1

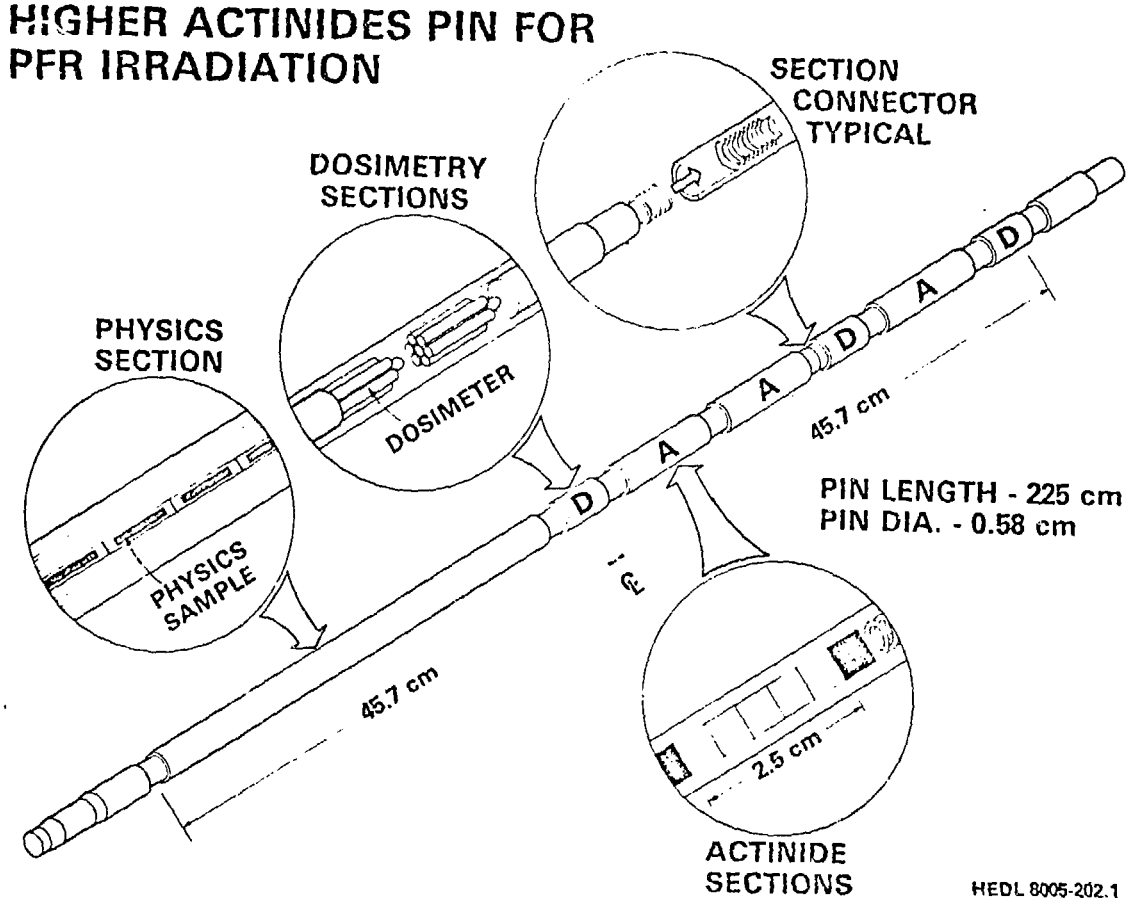
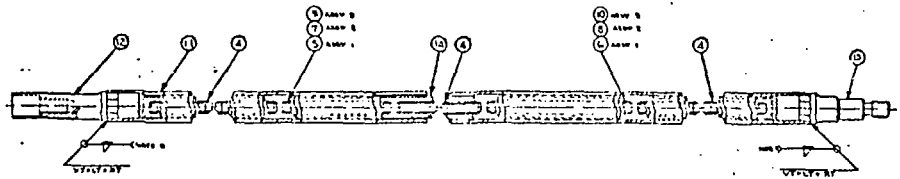


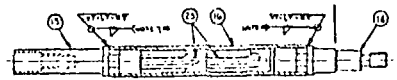
FIGURE 3. Higher Actinides Pin for PFR Irradiation. Neg 8006176-1cn

HIGHER ACTINIDES PIN SECTIONS

PHYSICS SECTION - 3 TOTAL



DOSIMETER SECTION - 13 TOTAL



ACTINIDE PELLET SECTION - 9 TOTAL

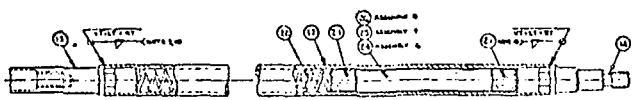


FIGURE 4. High Actinides Pin Sections.

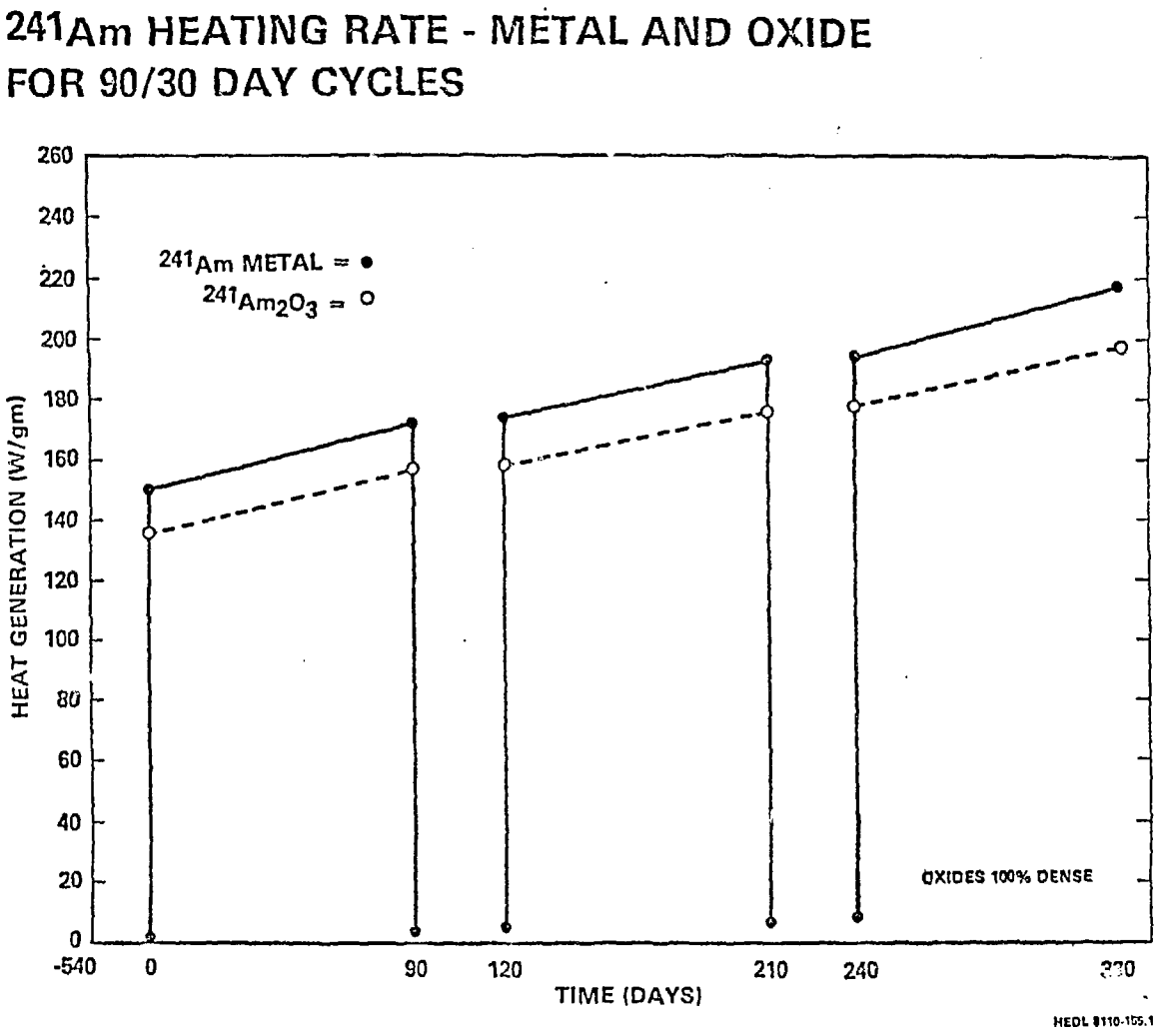


FIGURE 5. ^{241}Am Heating Rate - Metal and Oxide for 90/30 Day Cycles.

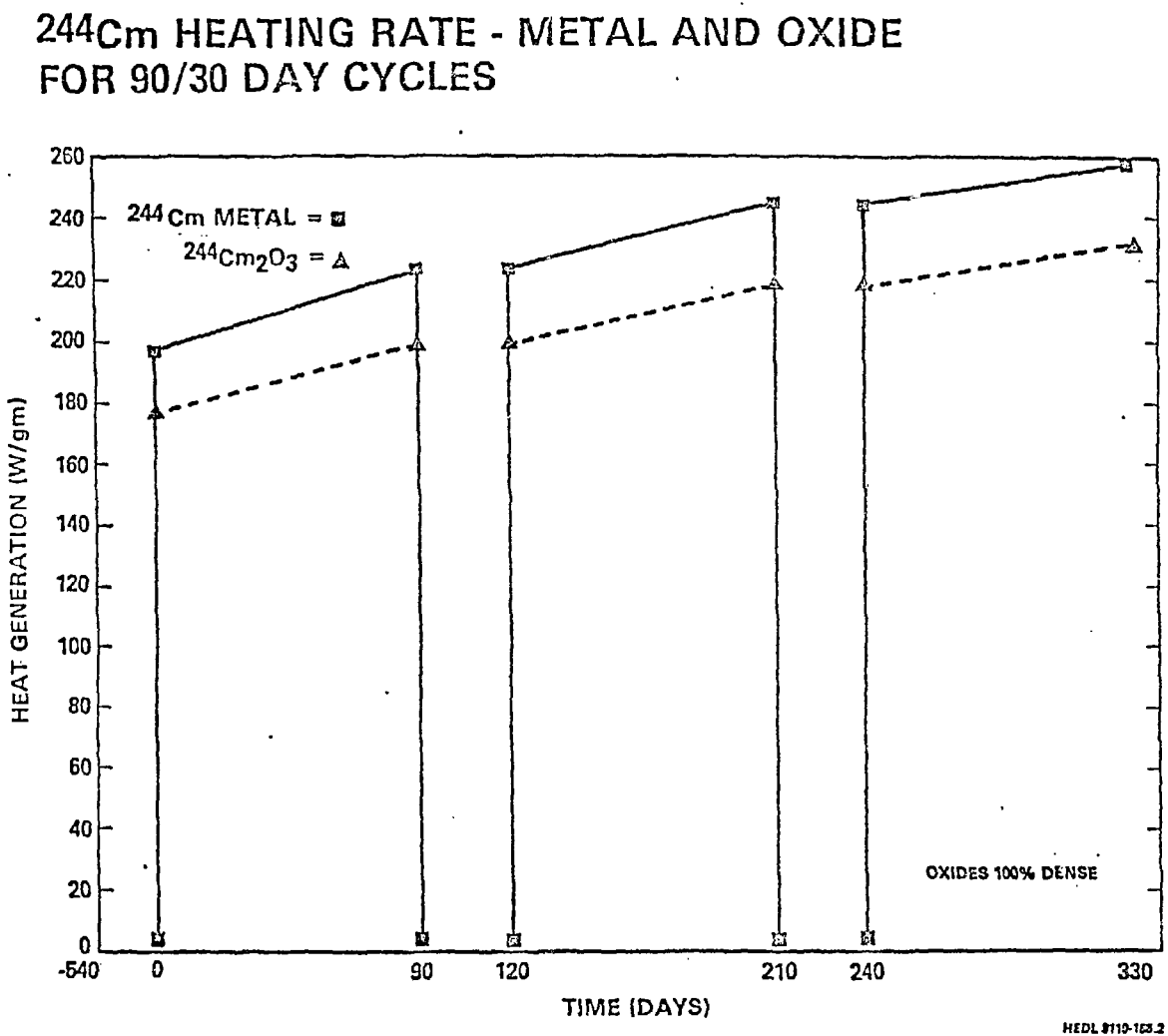


FIGURE 6. ^{244}Cm Heating Rate - Metal and Oxide for 90/30 Day Cycles.