SNE-101

**^{238}Pu PRODUCTION IN COMMERCIAL WATER REACTORS
EXPERIMENTAL AND ANALYTICAL PROGRAM**

February 1972

Southern Nuclear Engineering, Incorporated
Dunedin, Florida

MASTER

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PLUTONIUM-238 PRODUCTION
IN COMMERCIAL WATER REACTORS
EXPERIMENTAL AND ANALYTICAL PROGRAM

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I. INTRODUCTION AND SUMMARY

A study¹ performed by Southern Nuclear Engineering, Inc. in 1969 indicated that it might be feasible to produce plutonium-238 in significant quantities by irradiating neptunium-237 targets in commercial light water reactors. Confidence in the results of the study was limited by two factors: (1) the lack of experimental data on the behavior of neptunium targets exposed to a neutron spectrum characteristic of that in a light water reactor, and (2) the restrictions of the one-dimensional reactor physics model that was used.

In order to know what work would be necessary to determine better the feasibility of producing plutonium-238 in light water reactors, Southern Nuclear Engineering undertook in June, 1970 the project definition phase of a program to irradiate neptunium in an actual reactor and to generate a multi-dimensional reactor model for use in analyzing the results of the irradiation program. A detailed report² on the project definition phase was issued in September, 1970.

One task of the project definition phase involved contacting electric utilities that operated nuclear power plants and determining if there was industry interest in a cooperative program for the neptunium irradiation. Although all of the utilities contacted by Southern Nuclear Engineering (SNE) expressed interest in the proposed irradiation program, it was decided that

the best reactor for the program was the Connecticut Yankee reactor, owned by the Connecticut Yankee Atomic Power Company (CYAPC). CYAPC was quite interested in the program from the start and agreed to make space available for test specimens in its Connecticut Yankee reactor.

SNE began work on the program described here in October, 1970. The general objectives of the program were to obtain a no-cost agreement with CYAPC for the irradiation of Government-furnished neptunium-237 targets, to develop and prepare complete design drawings and specifications for the neptunium targets, to furnish guidance and assistance to Battelle Columbus Laboratories (BCL) with respect to the fabrication and shipment of the neptunium-237 targets, to select locations in the Connecticut Yankee reactor for the placement of the neptunium-237 targets, and to model both the Connecticut Yankee reactor and a later-generation pressurized water reactor in more than one dimension for subsequent calculations and analyses.

The USAEC approved the signed agreement between SNE and CYAPC on January 20, 1971. Approval of the design drawings and specifications for the neptunium-237 targets was received from the USAEC on December 3, 1970 and from both BCL and CYAPC on December 4, 1970. Prior to the approval of these drawings, a location was selected in the Connecticut Yankee reactor for the neptunium irradiation. The targets were fabricated at BCL. USAEC approval to ship the neptunium targets from BCL to the reactor site was received on March 17, 1971 and the targets were shipped

on that date. They were assembled at the Connecticut Yankee site and loaded into the assigned new-fuel assembly in accordance with the SNE drawings; SNE personnel were on-site during these operations.

It was learned on May 7, 1971 that the license amendment permitting irradiation of the targets in the Connecticut Yankee reactor had been received. A representative from SNE was at the site on May 7 and 8, 1971 to observe and monitor the loading of the new fuel assembly containing the neptunium targets into the reactor. Fuel loading was completed on May 8, 1971, and the reactor vessel head was re-installed on May 10, 1971. Hydrostatic tests were conducted on May 18, 1971, and low-power physics testing was completed on May 25, 1971, with full power attained on June 1, 1971. At the present time, the neptunium targets are undergoing irradiation and are scheduled to be removed from the reactor in June, 1972.

Concurrent with the effort leading to the neptunium irradiation, SNE worked toward obtaining a multi-dimensional model for the analysis of pressurized water reactors containing neptunium targets. An extensive review of multi-dimensional computer programs was conducted and, based on the review, the PDQ-7 program was chosen. The HARMONY program was selected to use in computing isotopic changes with burnup. The PDQ-7 and HARMONY programs are very complex, and their use in this project required considerable study. In addition to specifying and studying the multi-dimensional program and the isotopic-changes-with-burnup program,

it was necessary for SNE to work out the fundamental physics (including neutron slowing down, resonance capture, thermal spectrum, and thermal disadvantage factor methods) required for use with the programs. It was possible to modify some of the methods described in Ref. 1 and use them with the multi-dimensional models.

Calculations have been completed through the first core cycle for the Connecticut Yankee reactor, and both critical boron concentration with burnup and assembly-wise energy distribution at the end of life agree well with reported values. Work is in progress on modeling core 3 (the present core, which contains the neptunium targets) of the reactor for the purpose of following the irradiation of the neptunium targets.

The initial criticality and power distribution in two dimensions for the later-generation, or base, reactor considered in Ref. 1 have been calculated. Although no experimental data on this reactor will be available until it is operated, the results calculated by SNE agree well with values reported in the Final Safety Analysis Report for the reactor. Work will continue with the two-dimensional model of the base reactor to find the optimum arrangement for replacing burnable poison rods with neptunium targets.

II. AGREEMENT BETWEEN SOUTHERN NUCLEAR ENGINEERING, INC. AND CONNECTICUT YANKEE ATOMIC POWER COMPANY

During the project definition phase of the neptunium irradiation program, the USAEC requested that SNE obtain agreement from the participating utility on three points: (1) the utility would assume responsibility for any fuel assembly mechanical modifications necessary to accommodate the targets; (2) the utility would obtain any license approvals that might be required; and (3) the utility would continue to be responsible (liable) for the performance of its plant while the targets were being irradiated. CYAPC indicated from the start that it agreed to these three conditions.

A tentative agreement between SNE and CYAPC was drawn up by SNE's attorneys (Lowenstein and Newman, Washington, D. C.), and after many revisions and reviews by CYAPC, SNE, and the USAEC, the agreement was approved by the USAEC on January 20, 1971.

III. MECHANICAL DESIGN OF NEPTUNIUM TARGETS

The target rod assembly consists of two identical neptunium target rods which are mechanically assembled with, and suspended from, the base plate of a modified Connecticut Yankee flow-mixer device. The final configuration of the two target rods and flow mixer is illustrated by SNE Drawing No. 69-646-10 (Fig. 1). This drawing shows the installed position of the target rod assembly in the fuel assembly (Serial No. E-08) and also gives the core position (J-14) of the fuel assembly in Core III. The rotational orientation of target rods No. 1 and No. 2 is illustrated in Fig. 1. The J-14 fuel position is one of a number of positions which do not contain a rod cluster control (RCC) assembly. Specifically, the J-14 position is a higher-than-average-power "unrodded" fuel location which normally is fitted with a standard flow-mixer device; the flow-mixer device places stainless steel plugs (0.507-in. diameter by 5.30-in. long) in the upper end of the 20 unused (empty) RCC guide thimbles that are an integral, structural part of the fuel assembly. The relatively-tight-fitting plugs serve to minimize core coolant by-pass flow which enters from outside the thimble through six 1/8-in.-diameter flow holes located at the dashpot transition. The coolant flows up through the 0.512-in. thimble bore and leaves the bore at the top through the 0.003-in. plug-to-thimble annular clearance.

The mechanical design bases for the neptunium targets are such that the targets are expected to perform satisfactorily throughout the duration of their single-cycle test period. For normal full-power, steady-state operation, the reactor coolant nominal pressure is 2165 psia and its average temperature is 566^oF. The planned single-cycle test period for the targets extends from June 1, 1971 through June, 1972, and a cycle of this length at the assigned location is expected to result in an average fuel exposure of approximately 11,400 Mwd/mtU. The target rod loads, stresses, and strains resulting from the expected effects of reactor pressure, fission gas release, vibrations, and differential expansion during both steady-state and expected transient reactor operating conditions have been considered in the design of the target rod assembly and its support within the fuel-assembly RCC guide thimbles.

As shown in Figs. 1 and 2, each of the two identical target rods of the target-rod assembly contains four target segments which are designated as targets A, B, C, and D. Each target is basically the same design and consists of the required length of neptunium oxide pellets (0.385-in. diameter by approximately 1/2-in. long) encased in a Type 304 stainless steel target tube (0.422-in. O.D., with 0.0165-in. wall). An insulating zirconium oxide half-pellet is placed at each end of the spring-loaded pellet stack. A spring is located in the fission gas gap at the upper end and serves to hold all pellets in contact during handling and shipping of the target segments.

The target tube is seal-welded at each end after its final assembly with the required connector fittings. All target segments are filled with high-purity helium at zero gage pressure, to improve the pellet-to-clad thermal bond. Targets A, B, and C have a NpO_2 pellet stack length of nominally 10 in., while target D has an uninterrupted stack length of 43 in. The neptunium concentrations for the four types of targets are different, as specified in Table I of Fig. 1.

Target position detailed dimensions are presented in Fig. 1, with target A (highest concentration) located at the lower end of the target rod and with target D (lowest concentration) located at the upper end of each target rod. Generally, the four targets are arranged with the lower end of target A adjacent to the coolant inlet flow holes at the thimble-to-dashpot transition; then target B above, next target C with its mid-length centerline coinciding with the core mid-plane; finally target D with the upper end of its pellet stack coinciding with the upper end of the active core.

Typically, all targets are separated by a 5.750-in. inactive length which is required to provide the mechanical design features of Detail "A" in Fig. 1. Specifically, this "target dead band" includes such inactive components as (1) the two ZrO_2 half-pellets for insulation, (2) an extra-long-length connector-to-target tubing engagement for rod straightness control, (3) the fission gas gap for limiting pressure buildup and containing the compression spring, (4) the male-female threaded connection and locking

device for target joining, and (5) a generous length of the integral four-legged spacer fins for insuring the concentricity necessary for uniformity of coolant flow annulus.

The target tubing is nominally 0.422-in. O.D. by 0.389-in. I.D. (0.0165-in. wall) Type 304 stainless steel, which was cut from reactor grade fuel tubing stock furnished by Nuclear Materials and Equipment Corporation (NUMEC) at no cost (see Section IV). This tubing stock was from the production stock used in the manufacture of reload-fuel qualification assemblies for the Connecticut Yankee reactor. The detailed specification requirements for the tubing are contained in SNE Spec. No. 69-646-01 and are discussed in Section V of this report.

When inserted into the fuel assembly, as shown in Fig. 1, the two target rods are completely contained within the selected RCC guide thimbles at the designated locations. The vertical support of each target rod is provided at the upper end by its support rod and the bolted connection with the modified flow-mixer base plate, as shown in SNE Drawing 69-646-12 (Fig. 3). As illustrated in Fig. 1, the lower end fitting (0.444-in. diameter) is slipped into the upper end of the thimble dashpot bore (0.454-in. diameter) in a piston-cylinder type arrangement. Thus, the lower rod end is free to move axially as required by thermal expansion. Lateral support for the 9-ft length of the target rod is provided at five locations. Support at four of the locations is in the form of four-legged spacer fins integrally machined

into the upper connector of each target segment, with the fin tips contacting the thimble bore. The fifth lateral support point is provided by the slip-fit of the target A lower end fitting within the guide thimble dash bore.

Prior to insertion of the completed target-rod assembly into the fuel assembly, a pre-insertion inspection of the two receiving guide thimbles and dashpot bores is made. This inspection is made in accordance with Note 5 of SNE Drawing 69-646-10 (Fig. 1) and utilizes the thimble and dashpot inspection plug gages shown on SNE Drawing 69-646-13 (Fig. 4). The purpose of the inspection is to confirm that there will be no diametral interference upon insertion of the target rod assembly.

Referring to Fig. 2, target segments A, B, and C are mechanically assembled, locked, and shipped as the target rod lower section, while target D alone represents the target rod upper section. Each section is approximately 51 in. long. As a shipping convenience, the final assembly of the two lower sections, the two upper sections, and the modified flow-mixer assembly is performed at the reactor site and utilizes the assembly and upending fixture illustrated in Fig. 3. This fixture is also used for target insertion operations.

During the refueling outage in June of 1972, it is expected that the irradiated target rod assembly will be removed from the reactor while still contained within the fuel assembly. The fuel assembly containing the target rods will be transferred to a spent fuel pool storage rack for target

rod removal operations. Upon removal of the target rod assembly, the irradiated fuel assembly will be returned to the reactor for reassignment to a new position within the core for its second cycle of operation. Removal of the irradiated target-rod assembly will be performed under water, using the standard flow-mixing-device handling tool, which is routinely used to reposition flow mixers during fuel shuffling operations.

While the complete target rod assembly is vertically suspended under water from the flow mixer handling tool, the lower and upper sections of a target rod are inserted into their shipping position in the shipping cask basket. This basket is a removable insert which fits the internal cavity of the shielded shipping cask. The basket will provide four tube-type compartments, each having sufficient diameter and length to individually receive a 51-in.-long upper section or lower section of the irradiated target rod assembly. The basket is generally loaded as follows. The lower section of target rod No. 1 is manually inserted into the basket and the target rod is sheared by an underwater bolt cutter at the finned-spacer elevation of the threaded connector joint located between targets C and D. This sheared disconnect leaves the lower section (containing targets A, B, and C) in its basket-supported shipping position. The lower section of target rod No. 2 is then manually lowered into its shipping basket tube position and the second C-D connector joint is sheared by the bolt cutter. The target rod upper sections are then loaded into their shipping-basket tubes and are sheared

at the target D-to-support rod connector joint in a manner similar to that used for the lower sections. The entire basket is then lifted and lowered into the internal cavity of the shipping cask. The cask is closed and prepared for shipment to the target reprocessor.

IV. PRELIMINARY INFORMATION FOR LICENSING PURPOSES

In order to obtain the necessary amendment to the operating license for the Connecticut Yankee plant, CYAPC requested certain analyses of the expected behavior of the neptunium targets in the reactor. SNE performed the necessary calculations and, at the suggestion of CYAPC, also conducted flow tests on a mockup of the targets in a control rod thimble. Nuclear Materials and Equipment Corporation (NUMEC), fabricator of qualification assemblies for the Connecticut Yankee core, was contacted to determine if a control rod thimble and a fuel tube might be available to SNE for flow tests. NUMEC was extremely cooperative and loaned SNE a control rod thimble and a fuel tube for use in the tests. In addition, NUMEC agreed to give — at no cost — four fuel tubes to BCL to use as cladding for the actual neptunium targets.

A. Nuclear Calculations

Figures 5, 6, and 7 show, respectively, the calculated masses of Np-237 and Pu-238, and the estimated mass of Pu-239 as a function of average exposure of the fuel in the assembly containing the target. The Pu-239 masses are conservative; that is, they are high. These assumptions should make the calculations of heat generation rates quite conservative, since the largest contribution to the total fission cross section comes from Pu-239. The calculations did not include Pu-239 explicitly and, to be

on the safe side, it was assumed that the Pu-239 mass at any given exposure was equal to the original Np-237 mass minus the sum of the Np-237, Np-238, and Pu-238 masses at that exposure. In other words, the isotopes of plutonium beyond 239 were not considered. Further, losses of Pu-239 by fission were ignored. Figure 8 gives the power density in the target pellets as a function of fuel exposure. For the heat transfer calculations discussed later, the power densities were read from this graph at 12,000 Mwd/mtU — the expected exposure in the fuel assembly containing the targets. Figure 9 gives the total number of fissions in each target as a function of fuel exposure, and Fig. 10 gives the fission-gas pressure as a function of exposure. The fission-gas pressure was calculated as follows:

Using information from ORNL NSIC-37,³ and considering only the gases krypton, xenon, iodine, and bromine, it is seen that the total number of atoms of these gases produced per fission is 0.3113. When the total number of fissions for a given fuel exposure is multiplied by this number and divided by Avogadro's number, the result is the number of moles of gas, n . The pressure, P , is given by

$$P = n \frac{RT}{V}$$

where R is the gas constant, V is the free volume, and T is the absolute temperature.

Note that the pressures shown on Fig. 10 do not include the partial pressure of helium.

B. Thermal-Hydraulic Calculations and Tests

Thermal and hydraulic calculations were made and a few tests were conducted to determine what the thermal conditions would be in the target.

The criteria for acceptability were as follows:

- No bulk boiling shall occur in the target coolant at full reactor power or under reactor shutdown conditions;
- the target cladding tube outer surface temperature shall be at least 10°F below the normal, steady-state reactor coolant saturation temperature; and
- the $\text{NpO}_2\text{-ZrO}_2$ pellet centerline temperature shall not exceed 2500°F .

The above criteria were selected to insure that there would be no steam film blanketing of the target tube or any material damage to the pellets under any set of conditions.

A large fraction of the normal core coolant pressure loss occurs between the RCC thimble coolant inlet windows and the thimble coolant exit. The coolant velocity through a target thimble (v_t) can be approximated by

$$v_t = (v_f)(D_t/D_f)^{1/2}$$

where v_f is the coolant velocity in the surrounding fuel assembly, D_t is the hydraulic diameter of the target thimble annulus, and D_f is the hydraulic diameter of the fuel assembly. This approximate relationship will be shown to be applicable later, and it was used as a check on both the calculated and the experimental coolant flow data.

Data provided by CYAPC indicated a normal operating pressure difference between the thimble coolant inlet windows and the thimble coolant exit of 10.1 psi. Assuming an average coolant temperature (and using average temperature water properties) of 580°F, the target coolant flow rate was calculated by the common relationship

$$\Delta P = (K_i + K_s + K_o + \frac{f L}{D_t}) \times \frac{v^2 \rho}{2 g}$$

where K_i and K_o are the thimble inlet and outlet loss coefficients, respectively, and K_s is for the spacer loss. The $f L/D_t$ term dominates, and the flow rate is affected only slightly by the inlet and outlet restrictions and the spacers; therefore, a simple friction loss calculation is involved. Using the expression given above, a target coolant flow of 2.08 gpm was calculated. A value of about 2.0 results if the approximate expression for v_t given earlier is used. A flow of 1.4 gpm of 79°F water was measured in the flow tests at a pressure difference of 10.1 psi. Thus, the anticipated in-service target coolant flow was calculated by two methods, and the cold-water flow was determined experimentally as a check on the computational technique. Using cold water properties, the laboratory flow rates were predicted to within about $\pm 5\%$.

General temperature calculations were made for the target coolant, the target tube outer surface, and the target pellet centerlines. In addition, specific calculations were made to investigate the effects of off-centering of the target in the thimble on the local target tube surface temperature. The target coolant temperatures were calculated for an end-of-cycle, full-power situation. Reactor operating conditions assumed for the normal-operation calculations were as follows:

- Pressure at the top of the core is 2130 psia;
- the core coolant inlet temperature is 545°F ;
- the pressure difference between thimble coolant inlet windows and thimble coolant exits is 10.1 psi (excluding elevation difference); and
- the target coolant flow rate is 2.0 gpm.

The average heat transfer coefficient between the target and the coolant, calculated by the Dittus-Boelter basic equation,⁴ is $4300 \text{ Btu}/(\text{hr})(^{\circ}\text{F})(\text{sq ft})$. This value was used to calculate the outer surface temperature for both ends of each of the four target sections. A target pellet-to-tube "coefficient" of $500 \text{ Btu}/(\text{hr})(^{\circ}\text{F})(\text{sq ft})$ and a $\text{NpO}_2\text{-ZrO}_2$ thermal conductivity of $0.75 \text{ Btu}/(\text{hr})(^{\circ}\text{F})(\text{sq ft})/\text{ft}$ were used to determine the target centerline temperatures. A tabulation of the calculated temperatures is given in Table I.

Table I TARGET TEMPERATURES AT 12,000 Mwd/mtU

<u>Target</u>	Coolant temperature, °F		Target tube surface temperature, °F		Pellet centerline temperature, °F	
	<u>Bottom</u>	<u>Top</u>	<u>Bottom</u>	<u>Top</u>	<u>Bottom</u>	<u>Top</u>
A	557	568	583	594	1995	2006
B	568	578	591	601	1864	1874
C	578	585	594	601	1480	1487
D	585	603	595	613	726	744

Even though the target assembly is centered in the RCC thimble by spacers, consideration was given to the effects of eccentricity. If offset should occur locally, the coolant flow through the narrowest part of the annulus would be reduced; the departure from the average velocity would probably be a function of the local small-gap-to-average-gap ratio raised to the one-half power. If this assumption is made for a channel only half as wide as the average, the local coolant velocity is calculated, and the local heat-transfer coefficient is recalculated, the local target tube surface temperature increases only 10 to 25 °F. The first two criteria given earlier are not violated if these assumed conditions exist.

If the reactor coolant flow should be lost (or stopped) and the reactor shut down, the target power would decrease exponentially with time, as would the reactor core power. It can be shown that, even after loss of power to all four pumps, the core power would initially decrease faster

than the coolant flow. As core and target coolant flow will persist for more than 20 seconds, during which time the core and target powers will decrease to about 6% of their original levels, an analysis of this power level (6%) and zero core flow will determine the severity of loss of core coolant flow on target temperatures.

The distance between the thimble windows and the upper fuel assembly end is approximately 106 inches. It is this difference in elevation and the relative differences in coolant densities inside and outside the thimble that will tend to maintain target coolant flow under postulated zero flow conditions. (Actually, zero core flow cannot occur immediately after shutdown.)

As with all natural convection-circulation situations, the equilibrium flow-coolant temperature rise data must be determined by trial and error. However, for the situation here, the first iteration was based on the coolant entering at its normal average (inlet) temperature (545°F) and not being allowed to exceed the system normal saturation value (645°F minimum) less 10°F , specifically 635°F maximum. Under the shutdown conditions (6% power), a thimble flow of only about 0.06 gpm is required to prevent the coolant outlet temperature from exceeding 635°F . A pressure difference of about 0.01 psi is required to produce this flow rate. However, if the head resulting from the density difference between that for the allowable (here) target coolant average temperature and the average in the core is calculated, it is found to be approximately 0.20 psi, or many times that needed.

Under the loss of pumps condition, and with the core shutdown conditions of interest here, natural convection core coolant flow and heat transfer could obviously occur also. However, while this may reduce the relative coolant density differences, the core flow will provide a pressure difference and induce flow about the target.

During the manufacture of the oxide pellets for the targets, it was decided to run several thermal cycling tests to simulate the effect of reactor scram, which is the worst expected transient for the pellets. Maximum heat generation rates expected in the targets at 12,000 Mwd/mtU were used in a 12-radial-node model to calculate temperature changes within the pellets as a function of time following reactor shutdown. The heat generation rates were assumed to be uniform throughout the volume of the pellets. Figures 11, 12, and 13 show the radial temperature profiles as a function of time following reactor shutdown for Targets A, B, and C, respectively. The radial nodes in the heat transfer model are equally spaced and the heat generation rate is assumed to follow the ordinary decay heat curve for fission products. The bottom, horizontal line in each figure represents the sink (coolant) temperature, and the top line on each graph represents the center node temperature of the pellets. The center node temperature in each case was used by BCL as a guide for actual temperature transient tests in each type of pellet.

Flow tests were conducted in SNE's laboratory using a mill-run RCC thimble and a length of fuel tubing obtained from NUMEC. The tests were conducted by adjusting the water pressure at the thimble coolant inlet windows to a given value and then measuring the flow through the thimble-target tube annulus. The only differences between the test assembly and the target assembly under irradiation were the absence of target-section end fittings and spacers, and the lack of target pellets in the fuel tube. For the tests, the target (fuel tube) was filled with water for ballast.

A schematic of the flow-pressure drop test equipment is shown in Fig. 14. As previously stated, the inlet valve was adjusted until a pre-selected pressure was shown on a gauge (and hence a pre-set, but different, pressure differential reading was obtained), and the flow collected and measured over a timed interval. A high-precision gauge (whose calibration was checked) was used for the pressure determinations. Static pressure differences were confirmed from zero flow conditions.

A graph of pressure difference (corrected from test data) versus target coolant flow is shown in Fig. 15 for 79°F water. This graph can be used to determine the cold water flow through the assembly for any pressure difference in the range 4 to 23 psi. For the higher temperature reactor coolant, where both the viscosity and density are lower than for the test water, a higher volumetric flow will occur, as has been calculated.

The flow-pressure difference tests described above were run with the bottom end of the target tube centered by the RCC thimble dash-pot sections and the upper end by spacers. Upon completion of these "standard" tests, flow-annulus perturbations were effected intentionally, and additional data obtained. The dimensional perturbations were caused by (1) removing the upper end spacers and moving the target tube to first one and then the other most extreme offset position possible, and then (2) bowing the target tube inside the thimble such that there was contact between the two at some intermediate level. The latter bowing-offset was done for all points on the compass. These perturbations resulted in only barely perceptible flow changes, and the changes were always in the direction of increased flow. It was therefore concluded, as was expected, that target tube-to-thimble eccentricity would produce negligible coolant flow differences.

V. TARGET ROD ASSEMBLY SPECIFICATIONS

The detailed design, manufacturing, and final assembly features of the neptunium target rod assembly are given in the figures listed below and in SNE Specification No. 69-646-01 (Appendix A).

Target Rod Design Drawings

<u>Fig. No.</u>	<u>SNE Drawing No.</u>	<u>Title</u>
1	69-646-10, Rev. C	Neptunium target rod assembly in fuel assembly (J-14) of Conn. Yankee reactor
2	69-646-11, Rev. B	Neptunium target rod subassembly and details
3	69-646-12, Rev. B	Target rod flow mixer modifications and final assembly upending fixture
4	69-646-13	Target rod thimble plug gage

It should be noted that the design drawings and specifications can be considered as the documentation which defines the "as-built" final design specification. The requirements shown in the documentation evolved from the SNE design details and BCL manufacturing processes developed during the program. An example is the NpO_2 pellet specification, which was developed by BCL; it was patterned after the Yankee-Rowe fuel pellet specification and is given here in Appendix B. The target cladding specification is NUMEC Specification No. NPS-20-A, "Stainless Steel Cladding

Tubing," Revision 1; it is understood that this specification was used by NUMEC in procuring the cladding material for Connecticut Yankee qualification assemblies. Tubing from that procurement was generously donated by NUMEC for use as cladding material for the targets described here.

As stated above, Appendix A presents SNE Specification No. 69-646-01, "Neptunium Target Rod Assembly."

VI. MANUFACTURING PROCESS DEVELOPMENT AND FABRICATION OF TARGETS

This section presents the detailed results of the target rod manufacturing process development and fabrication phase of the program. This work was done exclusively by Battelle Columbus Laboratories. The BCL results include not only the developed manufacturing processes, welding, radiography, and leak-check procedures, but also product and component engineering tests and analyses. The test data include chemical analyses of pellet and clad materials, gas release data for various batches of NpO_2 pellets, mass spectrometric analysis of gas and moisture contents released from NpO_2 pellets at elevated temperatures, measurement of the thermal coefficient of expansion for NpO_2 pellets, pellet-to-clad compatibility tests at elevated temperatures, and the development of quality control procedures and check sheets.

The complete report of the BCL program on the neptunium target rod manufacturing process development and fabrication operations is contained in Appendix B of this report.

VII. ASSEMBLY OF THE NEPTUNIUM TARGETS AND THEIR INSERTION INTO THE FUEL ASSEMBLY

The neptunium target subassemblies were packaged and made ready for shipment from BCL on March 12, 1971. AEC approval to ship the targets was received on March 17, 1971. The targets were then transported to the Connecticut Yankee site, assembled, and loaded into the assigned new-fuel assembly in accordance with SNE Drawings 69-646-10 (Fig. 1) and 69-646-12 (Fig. 3).

Four radioactive subassemblies were transported from BCL to the reactor site. These subassemblies consisted of the two lower rod sections (targets A, B, and C) and two upper sections (target D), each 0.422 inch in diameter and approximately 51 inches long. The container used for the shipment is a Department of Transportation approved design that has the basic configuration of two 55-gallon drums welded end-to-end, with one end removable. The four target rod sections were sealed in plastic bags, wrapped with heavy aluminum foil (for beta shielding), and placed in the central, longitudinal cavity of the shipping container shielding media. The container was filled with a full-length stack of thick discs of pressed wood fiber shielding material. Each disc had a central hole about 3 inches in diameter, and these holes formed the central, longitudinal cavity. The bundle of target rod sections was loaded into the cavity with cushioning material at each end to minimize displacement during shipment. The

radiation levels at the surface of the loaded container were well below acceptable limits for the shipment.

Non-radioactive components (the modified flow mixer assembly) were packed and shipped in a conventional shipping carton.

The skid-mounted target rod shipping container, flow mixer carton, and necessary assembly tools and fixtures were transported by BCL personnel in a van-type truck from the Columbus Laboratories to the Connecticut Yankee site. There were no significant transportation problems or delays encountered in transit.

The shipment was received at the Connecticut Yankee site on March 17, 1971 and transferred to the fuel building for final assembly. Under direct control of the Connecticut Yankee reactor engineer and health physics personnel, BCL personnel removed the target-rod components from the shipping containers and performed the final assembly operation in accordance with SNE Drawing 69-646-10 (Fig. 1). This work was done on March 18, 1971. Later the same day, Connecticut Yankee personnel performed the operation of installing the completed target-rod assembly in its new fuel assembly (Serial No. E-08), in accordance with SNE Drawing 69-646-12 (Fig. 3). No significant problems or delays were encountered during these operations. SNE personnel were present during the target-rod final assembly and target installation in the fuel assembly. The fuel assembly containing the neptunium target rods remained in dry storage until it was installed in the reactor.

VIII. INSERTION OF THE FUEL ASSEMBLY INTO THE CONNECTICUT YANKEE REACTOR

On May 7-8, 1971, a representative from SNE was at the Connecticut Yankee site to observe and monitor the loading of the new fuel assembly (Serial No. E-08) containing the neptunium target rod assembly into the reactor. The fuel assembly had been moved from dry storage to the spent fuel pit, from which it was eventually moved to the reactor building. After being inspected visually for verification of serial number and orientation, the fuel assembly was lifted from the fuel transfer upender by the fuel manipulator crane and transported through the transfer canal to core position J-14 in Zone 3. The position is shown on SNE Drawing 69-646-10 (Fig. 1).

Connecticut Yankee personnel had indicated earlier their desire to perform an underwater target-rod partial withdrawal test (approximately a 3-ft lift) prior to the loading of the fuel assembly into the reactor. The withdrawal test was performed while the assembly was in wet storage, prior to its insertion into the core. The test verified the ability of the 30-ft-long flow mixer handling tool to grapple the modified flow mixer and simulated the required future operation of withdrawing the irradiated target-rod assembly from the partially-spent fuel assembly. SNE agreed the test was desirable, and advised Connecticut Yankee that there should be no problem in re-inserting the target rods subsequent to the partial

withdrawal test. SNE was later advised that the test had been successfully performed with no problems in removal or re-insertion.

Fuel loading was completed for core 3 on May 8, 1971. On May 10, 1971, the head was bolted on, and hydrostatic tests were conducted on May 18, 1971. Low-power physics testing was completed on May 25, 1971, and the reactor was taken to initial power on May 26, 1971. Full power was achieved on June 1, 1971. The neptunium targets are now in the operating reactor and will remain there until their scheduled removal in June, 1972.

IX. TWO- AND THREE-DIMENSIONAL COMPUTER MODELING

Work was begun in October, 1970 on the selection and writing of computer programs necessary to perform two-dimensional and three-dimensional analyses of a pressurized water reactor using Np-237 rods substituted for burnable poison rods. The model will be used to perform a number of two-dimensional calculations to determine the optimum arrangement for replacing burnable poison rods with Np-237 rods in the base reactor described in Ref. 1. The model will also be used to estimate neutron exposure, neptunium destruction, and Pu-238 production in the neptunium targets now in the Connecticut Yankee reactor. The calculations for the Connecticut Yankee reactor will be based on information received in the monthly operating reports from CYAPC.

Considerable time was spent in reviewing available two-dimensional diffusion theory computer programs in order to select one that was suitable for this work. As a result of the review, it was decided that the PDQ-7 program⁵ was the best available. HARMONY, a program⁶ which is coupled with and can be used with the PDQ-7 program to calculate isotopic changes with burnup, was studied extensively. Because of the apparent complexity of the HARMONY program, the coupling of a set of computer programs similar to those used in Ref. 1 with the PDQ-7 program was considered. After lengthy consultation with several experts on the PDQ-7-HARMONY

system, this idea was abandoned because the programming would have been extremely difficult and time consuming. Instead, in developing the computer model, procedures were set up to calculate fitted macroscopic and microscopic cross sections such that the reactor physics properties would be approximately the same as predicted by the methods described in Ref. 1.

The PDQ-7 program solves the neutron diffusion-depletion problem in one, two, and three dimensions. Up to five lethargy (or energy) groups are allowed, with the thermal neutrons represented by either a single group or a pair of overlapping groups. The geometries which can be considered are rectangular, cylindrical, spherical, and hexagonal. The first step in setting up the multi-dimensional model was to determine the minimum number of lethargy (or energy) groups necessary for an adequate representation of the reactor physics. The first clean core of the Connecticut Yankee reactor at time zero was chosen for a comparison of reactor physics parameters as a function of the number of energy groups considered in the calculations. Macroscopic diffusion equation coefficients for this reactor were calculated using the slowing-down, resonance capture, thermal spectrum, and thermal disadvantage factor methods described in Ref. 1. Two-dimensional (x,y) geometry was used and quarter-core symmetry was assumed. Diffusion equation coefficients averaged over three energy groups were used in one of the criticality calculations, while the other was based on the use

of only two energy groups. The same neutron spectra were used in calculating both the two-group and three-group coefficients. Calculated reactivities and assembly-wise power distributions for the two problems are shown in Fig. 16. A comparison of the two calculations indicated that two energy groups would be sufficient for the two-dimensional burnup model, thus saving considerable effort in cross section preparation, since microscopic values need to be prepared for only two groups instead of three.

Having established the group structure, the next item considered was that of representing the macroscopic cross sections (as calculated with the methods described in Ref. 1) as isotopic concentrations times microscopic cross sections for those isotopes which change with burnup plus macroscopic values for those isotopes which do not change with burnup. This representation is necessary in order to use the HARMONY program with the PDQ-7 program. For the purpose of making a comparison, a cell calculation on a pressurized water reactor of current design containing burnable poison rods was performed using the methods described in Ref. 1. This calculation essentially reduced the 38-group neutron spectrum to two groups and yielded the fast and thermal flux as well as the fast and thermal contribution to the multiplication constant. It also yielded the total multiplication constant. Several schemes for representing the microscopic and macroscopic cross sections for use in the HARMONY program were tried, and the simplest one seems to give answers that will be adequate

for the multi-dimensional analyses. The simple scheme simply averaged the time-zero microscopic cross sections for those isotopes whose concentrations change with burnup over two energy groups using the 38-group spectrum. The high energy group boundaries are from 0.625 ev to 10 Mev. The low energy group boundaries are from 0 to 0.625 ev. The only cross section which could not be handled in a microscopic way is the removal cross section. The procedure used to calculate the removal cross section was simply to calculate the macroscopic removal cross section as a function of burnup (U-235 concentration) with no soluble boron in the system, and then to apply a microscopic cross section to the soluble boron which corrected the total macroscopic removal cross section to the value calculated using the 38-group spectrum with soluble boron in the system. A comparison of the calculated neutron fluxes and k's as determined by the integral slowing-down thermal spectrum model and the microscopic model is shown in Table II. The agreement between the two methods as seen in Table II indicates that the microscopic model when used in the HARMONY program will give the same answers that would be obtained if the series of programs described in Ref. 1 were integrated with the PDQ-7 program for time-dependent analyses.

A flow chart of the calculations required for the two-group microscopic cross sections is given in Fig. 17. For each type of fuel assembly considered in the multi-dimensional model, the calculations described in the flow chart

Table II COMPARISON OF REACTOR PHYSICS PARAMETERS —
TWO-GROUP MICROSCOPIC VS. INTEGRAL
SLOWING DOWN — THERMAL SPECTRUM METHODS

Burnup Mwd/mtU	ϕ_1		ϕ_2		k_1		k_2		k_∞	
	<u>Integral</u>	<u>Micro- scopic</u>	<u>Integral</u>	<u>Micro- scopic</u>	<u>Integral</u>	<u>Micro- scopic</u>	<u>Integral</u>	<u>Micro- scopic</u>	<u>Integral</u>	<u>Micro- scopic</u>
0.0	33.67	33.65	5.938	5.937	0.2300	0.2303	0.7710	0.7704	1.001	1.001
1730.	33.70	33.68	5.766	5.728	0.2261	0.2262	0.7740	0.7761	1.000	1.002
3459.	33.71	33.69	5.673	5.615	0.2216	0.2217	0.7784	0.7817	1.000	1.003
13840.	33.80	33.71	5.863	5.704	0.1962	0.1960	0.8040	0.8072	1.000	1.003
15290.	33.81	33.69	5.955	5.789	0.1926	0.1921	0.8076	0.8104	1.000	1.002

are performed as a function of burnup over the range of burnup expected in that type of fuel assembly. These calculations are performed with no soluble boron in the moderator. In order to deduce the microscopic removal cross section for soluble boron, a time-zero calculation including a reasonable soluble boron concentration is performed and the microscopic removal cross section for soluble boron is then defined as the boron concentration times the microscopic removal cross section for boron plus the macroscopic removal cross section for the system with no soluble boron considered. The two-group microscopic cross sections from the time-zero calculation are used throughout the burnup, and self-shielding factors which are tabulated as a function of concentrations in the HARMONY program are used to modify appropriate cross sections when necessary.

HARMONY is a very generalized program. The primary emphasis in the design of the HARMONY system has been to obtain a high degree of flexibility and increase the range of problems that may be described. In the past, it has been difficult to use depletion programs for a class of reactor for which the program was not specifically designed. HARMONY allows the user to formulate a depletion program to meet his needs. Rather than tailoring his reactor to fit the requirements of a particular depletion program, the user is able to tailor the depletion system to describe his reactor. Figure 18 shows the neutronic chains SNE has used in HARMONY for the present work. Calculations utilizing these chains

have been compared with calculations using analytic solutions to the differential equations describing the chains and agreement is excellent.

A complete two-dimensional calculation of the first core in the Connecticut Yankee reactor was performed using the PDQ-7—HARMONY program. The assembly-wise relative power density as a function of average core burnup is shown in Fig. 19. Figure 20 gives the assembly-wise burnup at a core average burnup of 16,000 Mwd/mtU; the burnups were calculated from the fission product inventory remaining in each fuel assembly. Comparison of the data given in Fig. 20 with predicted and calculated values given in the Connecticut Yankee operating report for June, 1970 shows that the SNE calculations are in reasonable agreement with reported values.

Figure 21 shows the calculated versus actual critical boron concentrations as a function of burnup for the Connecticut Yankee reactor. The reported values were taken from the monthly operating reports, and the calculated values are based on a point model using methods described in SNE-53. Boron concentrations calculated with the point model are used directly in the PDQ-7 — HARMONY calculations. Over the entire cycle, k_{eff} is 1.0 to an accuracy of better than 0.5% in the PDQ-7 — HARMONY calculations. The agreement obtained between reported and calculated values of critical boron concentrations and assembly-wise burnups indicates

that the model is adequate to analyze the irradiation test and to perform the two-dimensional parametric study.

The Consolidated Edison Company of New York kindly agreed to furnish specifications for the first core of the Indian Point Unit No. 2 reactor (the base reactor to be used in the parametric study), including all information on burnable poison rods and their location in the core. Following receipt of this information, work was begun on setting up a two-dimensional calculation of the Indian Point 2 core. The two-dimensional power distribution at the beginning of life has been completed and is shown in Fig. 22. The figure shows, in addition to SNE calculations of the power distribution at the beginning of life, the values reported in the Final Safety Analysis Report. It can be seen that both the vendor's calculations and Southern Nuclear's calculations predict the same fuel assembly will have the maximum relative power density and that the other power distribution numbers are in reasonable agreement. Work is continuing on the burnup of this system and the results of the burnup through the first core cycle will be reported in FY 72.

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APPENDIX A

SNE DESIGN SPECIFICATION NO. 69-646-01

NEPTUNIUM TARGET ROD ASSEMBLY

JUNE 30, 1971

APPENDIX A

SNE DESIGN SPECIFICATION NO. 69-646-01, NEPTUNIUM TARGET ROD ASSEMBLY, JUNE 30, 1971

1.0 Scope

- 1.1 This specification defines the design requirements for manufacture and assembly of the neptunium target rod assembly to be irradiated in Core III of the Connecticut Yankee reactor.

2.0 Design

- 2.1 The fabrication of the target rod assembly shall be in accordance with this specification and the latest revision of SNE Drawings Nos. 69-646-10, 69-646-11, 69-646-12, and 69-646-13.
- 2.2 No change shall be made in the detailed design requirements or quality of material furnished under this specification without the written approval of Southern Nuclear Engineering (SNE).

3.0 Components

3.1 Target Rod Pellet Specification

- 3.1.1 The requirements of the neptunium dioxide target rod pellets are contained in Attachment No. 1 to this specification. The detailed pellet specification was developed by Battelle Columbus Laboratories (BCL) as part of the target rod fabrication process development program in order to satisfy the SNE pellet design requirements. The development of the NpO_2 pellet specification is discussed in detail in a BCL report.*
- 3.1.2 A minimum of two new pellet samples for each NpO_2 concentration and one sample of each machined component shall be available to SNE for post-test evaluation.

* See Appendix B of this report.

3.2 Target Rod Tubing Specification

The target rod tubing was obtained from NUMEC and was supplied to this program from their stock of seamless Type 304 stainless steel tubing (0.422 in. O.D. by 0.0165 in. wall) used as reactor-grade fuel rod tubing during NUMEC fabrication of Connecticut Yankee qualification assemblies. This "as received" tubing was manufactured per the requirements of the NUMEC Purchasing Specification NPS-20-A, Revision 1, and therefore these details are applicable to the definition of target tubing used in the neptunium target rods installed in the Connecticut Yankee reactor Core III. The NUMEC specification was reviewed by SNE and was judged to be satisfactory for the application of target tubing. The NUMEC Purchasing Specification is included as Attachment No. 2 of this Specification.

3.3 All Other Components

3.3.1 The materials shall be as specified on the drawings.

3.3.2 Complete certification of each material, including mill test reports to the applicable specifications, must be provided to SNE prior to manufacture of components. All data obtained during the performance of required testing shall be included. Mill or manufacturer's test reports shall contain actual test results of chemical and physical analyses performed on the raw materials.

4.0 Component Identification

4.1 All material shall be carefully identified and segregated from other materials in the vendor shop. No substitutions shall be made.

4.2 Records shall be maintained by the vendor identifying each type or batch of manufactured parts.

5.0 Procedures

5.1 Welding

Welding of connector end fittings to target tubing shall be performed to totally contain the solid and gaseous components of the target segment. Parameters utilized in the welding will be established prior to target segment fabrication by practice set-up and destructive analysis. Operators for the welding process will be qualified on the basis of practice welds.

5.2 Loading

To insure that all details described in SNE Drawing No. 69-646-11 are fully met during loading and assembly of the target rod segments, readings from gauges used throughout this effort will be recorded as evidence that the work has been completed according to specifications.

5.3 Decontamination

Radioactive contamination on the outside of the rod is removed so that the target can be safely handled outside the glove boxes for final inspection and assembly.

5.4 Visual Examination

The target rod segment will be visually examined for defects such as dents, scratches, etc., which might have been incurred during handling for loading and welding.

5.5 Leak Check

Each target rod segment will be leak checked to assure the quality and integrity of the weldments and cladding. Two types of leak checks will be performed: (1) element maintained under high pressure helium and then immersed in liquid for observation of bubble formation, and (2) element with helium plenum gas maintained under vacuum and presence of helium analyzed with a CEC mass spectrometer leak detector with a sensitivity of 1×10^{-9} std cc/sec of He for full-scale deflection of the leak indicator meter.

5.6 Radiography

All weld closures will be radiographed by X-ray techniques in accordance with ASTM Standard E94-62T. The exposure and quality of the radiographic prints shall be such that weld porosity and weld thickness may be determined.

6.0 Cleanliness

- 6.1 The final part, ready for shipment, shall be free of all foreign matter and contamination.
- 6.2 Clean methyl alcohol or a specifically approved alternate shall be used to wash and rinse the finished parts just prior to packaging.

7.0 Quality Control and Inspection

- 7.1 The vendor's quality control inspection program is subject to review by SNE.
- 7.2 All inspection and testing methods shall be subject to review and approval by SNE during production. Corrective action shall be taken by the vendor in any instance where inspection and testing methods are inadequate to assure that components are meeting specification.
- 7.3 SNE shall have reasonable access to manufacturer's plant during the fabrication and testing of all parts. The vendor shall provide reasonable assistance to the SNE representative in his duty to assure compliance to this specification and its drawings.
- 7.4 Specific approval by SNE is required prior to any shipment of manufactured parts to the Connecticut Yankee site.
- 7.5 Adequate records shall be maintained throughout all stages of inspection (receiving, in-process, final, and test). The inspection records format shall be the vendor's choice to assure data are properly recorded.

8.0 Preparations for Shipment

- 8.1 Unless otherwise specified, all components are to be individually packaged in a polyethylene bag or sleeving. The ends shall be

heat sealed or stapled. Alternate packaging methods and materials may be submitted to SNE for approval.

- 8.2 A suitable shipping container shall be provided with sufficient shielding to attenuate the target rod radiation to a safe level at contact with the container outer surface.

9.0 Surface Contamination

- 9.1 All target rod outer surfaces shall be free of ferrites. No carbon steel or iron containers or handling tools shall be used after the components are machined. Stainless steel or an approved plastic container only shall be used to hold the parts prior to packaging for shipment.
- 9.2 Halogen compounds, acids of a pH less than 6 and bases of a pH greater than 8 and boron compounds are considered detrimental and shall not be used. Approval is granted to use acetone to remove grease, but it must be followed by a methyl alcohol rinse. The use of any other materials must be specifically approved by SNE.
- 9.3 The surface of the finished parts shall be free of dirt, grease, dust, chips, lubricants, residues by fixtures, handling tools, containers or other materials.

TARGET ROD PELLETT SPECIFICATION

1.0 Scope

- 1.1 This specification applies to solid cylindrical NpO_2 -(CaO - ZrO_2) target pellets that will be encapsulated and irradiated in the Connecticut Yankee reactor Core III of the Connecticut Yankee Atomic Power Company (CYAPC).

2.0 Design Requirements

- 2.1 No change shall be made in the design details or quality of material furnished under this specification without the written approval of Southern Nuclear Engineering (SNE).

3.0 Manufacture

- 3.1 The pellets shall be prepared from neptunium dioxide (NpO_2) powder supplied by Savannah River and reactor grade calcia (CaO) stabilized zirconia (ZrO_2) containing 10-12% CaO purchased from ZIRCOA. Standard cold pressing and sintering techniques will be used for all pellet fabrication. No materials shall be added to the powders except small quantities of organic binders and/or an organic lubricant needed for pelletization.

4.0 Materials Accountability

Accountability and accounting for the NpO_2 would be in accordance with AEC Manual Chapter 7400. Battelle Columbus Laboratory (BCL) is an exempt AEC contractor operating under the contract cognizant of the Chicago Operations Office. Battelle Columbus will be responsible to the AEC through the Chicago office for the receipt, processing, removals, and losses of the NpO_2 through established accountability and accounting procedures. At the conclusion of the program, a closed accountability balance of all materials received will be furnished to both SNE and the AEC. The disposition of any generated scrap or unused materials will be in accordance with instructions furnished by SNE.

5.0 Chemical Characterization and Tests

- 5.1 Neptunium contents of the pellets for the various target segments will conform to those values specified below, which are based on the information given in SNE Drawing No. 69-646-10.

Segment		Np density g/cm ³	Total volume of fuel segment, cm ³	Total mass Np, g	Total mass NpO ₂ g	NpO ₂ composition of target pellets, wt. %
1A	2A	5.44	18.88	102.69	116.55	77.22
1B	2B	2.72	18.88	51.35	58.28	48.76
1C	2C	1.36	18.88	25.67	29.13	28.06
1D	2D	0.68	81.17	55.20	62.65	15.18

- 5.2 Chemical analysis for Np, Zr, and Ca will be performed in accordance with established procedures for these elements.
- 5.3 Since the oxygen-to-metal ratio of the pellets is unknown and would be meaningless unless referenced to back-up data on phase relationships and stoichiometry, emphasis for pellet characterization and behavior will be placed in microstructural examinations and prolonged heat treatment of encapsulated pellets. Stoichiometry of the pellets will be that which results from sintering to high temperatures in an atmosphere of high purity nitrogen and is expected to vary slightly for each composition because of the changes in NpO₂ and ZrO₂ contents.
- 5.4 Impurities present in the pellets shall not exceed the levels given below:

<u>Impurity element</u>	<u>Maximum content, ppm</u>	<u>Impurity element</u>	<u>Maximum content, ppm</u>
Ag	0.3	Mn	150.0
Bi	2.0	Mo	50.0
B	1.0	Ni	100.0
Cd	5.0	Pb	60.0
Cr	200.0	Si	300.0
Cu	25.0	Sn	40.0
Fe	500.0	V	20.0
Li	0.5	Zn	50.0
Mg	300.0		

- 5.5 Spectrographic analysis will be utilized to determine trace impurities.
- 5.6 The hydrogen content of the pellets shall not exceed 10 ppm, including the amount contributed by water. Hydrogen content shall be determined from mass spectrographic analysis of a gas sample taken during measurements for total gas in 5.9, below. It shall be assumed that the ratio of hydrogen to other species found in this gas sample is for all practical purposes the same and this ratio will be used to calculate the hydrogen content of pellets analyzed for total gas release.
- 5.7 Chlorine content shall not exceed 10 ppm as determined from an analysis of a pellet randomly selected from a batch of pellets containing 15.18 wt. % NpO_2 .
- 5.8 Total water content shall not exceed 10 ppm. Moisture desorption temperatures employed in the analysis shall be 1000°C .
- 5.9 Total gas release inclusive of water shall not exceed 0.05 cc/g of target material at S. T. P. Gas release shall be measured by out-gassing for at least 30 minutes at 1×10^{-6} mm Hg at a temperature of 1000°C .
- 5.10 Pellets of each composition will be sectioned and examined metallographically to magnifications of 500 X. This examination shall indicate a minimum of 90% solid-solution of the NpO_2 calcia stabilized ZrO_2 powders.
- 5.11 Pellets examined metallographically will be subjected to quantitative alpha autoradiography. The uniformity of the neptunium dispersion shall be such that there are no areas of high NpO_2 concentration exceeding 200μ .
- 5.12 One pellet of each composition will be encapsulated in a Type 304 stainless steel can and heat treated for one month at a temperature of 500°C . The cladding and pellets will be examined for diffusion interaction of the various constituents prior to the loading of the targets into the reactor.

6.0 Physical Properties and Dimensions

- 6.1 Densities of finished pellets shall be in the range specified below for each target composition.

<u>Segment</u>		<u>NpO₂ composition,</u> <u>wt. %</u>	<u>Density,</u> <u>g/cm³</u>
1A	2A	77.22	8.60 $\pm$ 0.15
1B	2B	48.76	6.10 $\pm$ 0.15
1C	2C	28.06	5.40 $\pm$ 0.15
1D	2D	15.18	5.10 $\pm$ 0.15

- 6.2 Three pellets shall be selected from the pellets prepared from each batch of material and measured for weight and volume. Geometric densities determined from these measurements shall fall within the range specified in 6.1 above.
- 6.3 Pellet dimensions shall conform to those given on SNE Drawing No. 69-646-11. Tolerances on the pellets are 0.380 to 0.385 inches for the diameter and 0.350 to 0.650 inches for the length.
- 6.4 The plane of each pellet end shall not deviate from a plane perpendicular to the center axis by more than 0.08 inch across the pellet diameter.
- 6.5 The pellets shall be subjected to visual inspection for chips and fissures and they shall conform to the following specifications.
- 6.5.1 Chipped pellets shall not have lost more than 10% of the area at either end.
- 6.5.2 The sum of the circumferential length and depth of each imperfection on the cylindrical surface shall not exceed 0.10 inch. The maximum axial length of any one chip shall not exceed 0.10 inch. The sum of the circumferential lengths of all chips and pock marks in any one plane perpendicular to the pellet axis shall not exceed 0.10 inch. In cases of "borderline" acceptability, chipped or fissured pellets may be accepted when they are within the weight per unit volume specification and withstand subsequent handling and tube loading without further deterioration.

7.0 Test Sampling

7.1 Dimensional Samples

- 7.1.1 Each pellet shall be inspected visually for chips, fissures, and other imperfections.
- 7.1.2 All pellets shall be inspected for compliance to dimensional tolerances as stated in 6.3.
- 7.1.3 For pellet squareness, a portion of the pellets may be randomly sampled. The sampling plan and rejection criteria shall be subject to mutual agreement.

7.2 Chemical Samples

- 7.2.1 A batch of material shall be defined as the NpO_2 and CaO stabilized ZrO_2 powders blended initially by wet ball milling in the amounts necessary to obtain the NpO_2 compositions specified in 5.1. A batch of pellets shall consist of the compacts pressed and sintered under identical conditions from a batch of material as defined above.
- 7.2.2 One pellet prepared from each batch of material shall be selected and stored as an archive sample to be utilized during post-irradiation evaluation of the target rods.
- 7.2.3 An analysis is to be performed on one pellet from each batch of material to assure compliance with Section 5.0.
- 7.2.4 Gas release will be determined using an entire pellet selected at random in 7.2.3
- 7.2.5 Sections from one pellet from each pellet batch will be examined metallographically and submitted for quantitative autoradiography.
- 7.2.6 One pellet of each composition will be encapsulated in a Type 304 stainless steel can and heat treated for one month at 500°C .

8.0 Test Reports

- 8.1 Copies of the chemical analyses reported for total Np, Zr, Ca and impurities together with the analyses taken on the cladding and end component material plus reports of physical properties and dimensions shall be submitted to SNE and CYAPC on completion of the program.

9.0 Packaging and Handling

- 9.1 The pellets shall be packed and handled in a manner that will insure their compliance with the above specification when loaded into the cladding tubes.
- 9.2 The identity of pellet batches shall be retained throughout the manufacture of finished target rod assemblies.

10.0 Inspection

- 10.1 The manufacturer shall afford SNE and CYAPC representatives all reasonable facilities, information, and right of inspection to satisfy themselves that the pellets furnished are in accordance with this specification.
- 10.2 Acceptance of pellets and target rods by SNE or CYAPC shall release BCL of all responsibility for performance of these rods while in the reactor.

11.0 Compliance with Regulations

- 11.1 The manufacturer shall have appropriate licenses and clearances required for compliance with all applicable regulations of Federal, State and Local bodies with respect to receiving, accounting for, processing, storing and shipping neptunium.

NUMEC PURCHASING SPECIFICATION

NPS-20-A

Revision 1

July 11, 1969

STAINLESS STEEL CLADDING TUBING SPECIFICATION

1. Scope

- 1.1 This specification details the requirements for seamless stainless steel tubing for fuel rod application.

2. References

- 2 1 The latest revisions of the following supplementary specifications are part of this specification. In the event there is a conflict between the standard and the latest revision thereof, the manufacturer shall notify NUMEC in writing. The latest revision shall take precedence unless NUMEC advises otherwise within five days after being notified of the discrepancy.

2.1.1 Technical Society Standards

- 2.1.1.1 ASTM-A-213-66 Specification for Seamless Ferritic and Austenitic Alloy - Steel Boiler, Superheater, and Heat-Exchanger Tubes.
- 2.1.1.2 ASTM-A-370 Methods and Definitions for the Mechanical Testing of Steel Products.
- 2.1.1.3 ASTM-E-112 Estimating the Average Grain Size of Metals.
- 2.1.1.4 ASTM-A-393 Recommended Practice for Conducting Acidified Copper Sulfate Tests for Intergranular Attack in Stainless Steel.
- 2.1.1.5 Mil-Std-271 Non-Destructive Testing Requirements For Metals.

3 Fabrication Process

- 3.1 Stainless steel shall be made by the electric furnace process. The tubing shall be cold worked and tempered. Following the solution anneal a maximum of 15% cold work shall be permitted. Following the temper point anneal the material shall not be heat treated in any manner. The completeness of the final solution anneal shall be verified by microscopic examination. No continuous carbide precipitation network is permitted in the grain boundary.

4 Material

- 4.1 The tubing shall be seamless Type 304 Stainless Steel as per ASTM-A-213-66 except where this specification has imposed additional requirements and/or tests.

5. Lot Size

- 5.1 A lot shall consist of all material of the same size finished from the same ingot and annealed in the same final vacuum annealing charge. A lot shall consist of units which are successively produced by the same manufacturing processes at essentially the same time. The lot size shall not exceed 2500 feet for tubes one inch or less in outside diameter and shall not exceed 1500 feet for tubes over one inch but less than two inches in outside diameter.

6 Product Requirements

6.1 Chemical Composition

- 6.1.1 Chemical composition limits shall be as per ASTM-A-213-66 with the following modifications:

Ni	9.0 to 11.0%
Cr	17.0 to 19.0%
Co	.10% Max. (As close to .05% as possible)
B	.003% Max.

- 6.1.2 Two check analysis shall be made on tubes produced from each heat.

6.2 Mechanical Characteristics

- 6.2.1 Properties: The tubing shall have the following properties:

6.2.1.1 Ultimate Strength

Room Temperature	100,000 psi, min.
650°F	70,000 psi, min.

6.2.1.2 Yield Strength, 0.2% OFFSET

Room Temperature	75,000 psi, min.
650°F	60,000 psi, min.

6.2.1.3 Ductility, Elongation in Two Inches

Room Temperature	35% min.
------------------	----------

6.2 1.4 Grain Size

Two metallographic specimens, one longitudinal and one transverse section, shall be selected at random from each lot for grain size determination. The maximum grain size shall be ASTM Micro-Grain Size Number 70, as specified in ASTM-E-112, "Estimating the Average Grain Size of Metals".

If any sample fails the metallography test, the lot shall be retested using five samples taken at random. If any of the five samples fail, a sample of every tube in the lot shall be tested. Any tube which fails to meet the grain size shall be rejected.

6.2 1.5 Proof Test

6.2.1.5.1 The required hydrostatic proof test of ASTM-A-213 is waived.

6.2.1.5.2 A minimum of one sample 12 inches in length or greater from each lot shall be burst tested to failure at 650°F. Where either the pressure or circumferential elongation at failure is less than required, two additional samples from the same lot shall be tested in the same manner. If failure of either sample occurs, the entire lot shall be rejected.

Burst Pressure

The minimum burst pressure shall be calculated using the following formula:

$$P = \frac{2ST}{D}$$

Where:

S = Min. specified tensile strength (psi) at 650°F.

D = Nominal inside diameter + 2 x minimum wall thickness (inches),

t = Minimum specified wall thickness (inches).

Circumferential Elongation

The minimum circumferential elongation shall be 16 percent. The elongation shall be calculated as follows:

$$\% \text{ Elong.} = \frac{C_f - C_o}{C_o} \times 100$$

Where:

C_o = Original circumference = (nominal outside diameter).

C_f = Final circumference to be measured on the fractured burst test specimen (This measurement does not include the distance between the fracture faces).

6.2.1.6 Flattening Test

6.2.1.6.1 One 3 inch section of tubing from each end of one finished tube from each lot shall be flattened at a uniform rate to a thickness of 3 times the nominal wall thickness in accordance with ASTM-A-370. The flattened tubing shall show no cracking or rupture visible to the unaided eye.

6.3 Physical Characteristics

6.3.1 Dimensions: The tubing shall comply with the dimensions and tolerances of the applicable NUMEC drawing.

6.3.1.1 Wall Thickness: Wall thickness shall be measured on all tubing using Vidigage or an equivalent technique acceptable to NUMEC. The method shall provide for at least one continuous 360° inspection of the wall thickness every 6 inches of tubing length, and may be checked concurrently with ultrasonic testing.

6.3.1.2 Inside Diameter: Inside diameter shall be measured on all tubing using a two orifice air gauge or an equivalent method acceptable to NUMEC. The method shall provide for at least one continuous 360° inspection of tubing inside diameter every 12 inches of length.

6.3.1.3 Ovality: Ovality shall be measured by a method acceptable to NUMEC

6.3.1.4 Surface Finish: Surface finish shall be checked by 100% visual inspection of all tubing. Questionable areas shall be checked with a Profilometer.

6.3.1.5 Defects: Tubing shall be free of burrs, tears, grooves, laminations, slivers or foreign bodies, pits, grinding marks and internal and external imperfections such as handling marks, dents, die, roll, or mandrel marks. In local areas from which metal is removed, displaced, or absent the depth shall not exceed 10% of the nominal wall thickness. Compliance with the defect requirements shall be established by the following methods:

- 6.3.1.5.1 Ultrasonic inspection of all tubing with techniques capable of detecting defects equivalent to 10% of the wall in depth, .004" in width and .060" long. Calibration standards for setting up the equipment shall be prepared from a length of the finished product. A change in processing conditions shall require a change in standards. The standards shall contain 2 O.D., and 2 I.D. notches, one transverse and one longitudinal in each instance.
- 6.3.1.5.2 Fluorescent penetrant inspection of the outside tubing surfaces in accordance with Mil-Std-271 using Type V low chloride material. A sufficient number of samples shall be taken to assure a 95% x 95% confidence level on the tubing.
- 6.3.1.5.3 100% visual inspection without magnification.
- 6.3.1.5.4 One tube per lot shall be sectioned and the inner surfaces examined for defects and surface finish.

6.4 Cleanliness: The inside and outside surfaces of the tubing shall not be contaminated by grease, dirt, metal particles and other foreign matter. Residual halide contamination shall not exceed .0005 mg/in².

6.5 Corrosion: Two representative samples per lot shall be corrosion test in accordance with ASTM-A-393. The specimens shall be free of intergranular corrosion.

7. Tests and Inspection

7.1 Methods of inspection to assure compliance with all applicable drawing, specification and purchase order requirement shall be specified by the supplier and submitted to NUMEC for approval.

- 7.2 All visual inspection shall be made without magnification under a minimum of 125 foot candle illumination.
- 7.3 Materials used for manufacture of the tubing shall be certified by the manufacturer. Results of all raw material tests shall be included in the certification package, including mill test reports.
- 7.4 Measurement accuracy shall be established by using instrumentation having been calibrated to 10% of the specification tolerance.

8 Quality Control

- 8.1 Quality Control procedures, sampling plans, inspection methods and equipment shall be subject to NUMEC approval.
- 8.2 The Manufacturer shall submit a general process outline for NUMEC approval.
- 8.3 The manufacture, testing and inspection of the tubes shall be subject to surveillance by NUMEC's Quality Control Department. The purchaser shall allow reasonable access to the areas involved by NUMEC inspectors in order to verify compliance with applicable requirements.
- 8.4 All product is to be released by NUMEC Quality Control to shipment.

9. Packaging

- 9.1 Tubes shall be packaged in a manner to prevent damage in shipment. Tubes shall be individually packaged in paper sleeves or the ends capped to prevent contamination of the inside surfaces.

APPENDIX B

MANUFACTURING PROCESS DEVELOPMENT
AND FABRICATION OF
NEPTUNIUM TARGET RODS

BY

BATTELLE COLUMBUS LABORATORIES
COLUMBUS, OHIO

PREPARATION AND CHARACTERIZATION OF
 $\text{NpO}_2\text{-CaO-ZrO}_2$ TARGET RODS FOR Pu^{238} PRODUCTION
IN COMMERCIAL WATER REACTORS

by

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PREPARATION AND CHARACTERIZATION OF
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SUMMARY

Two neptunia-zirconia-fueled target rods, each containing four sections of different neptunia concentrations, were fabricated and loaded into Connecticut Yankee Reactor* for irradiation. Nominal compositions of the target pellets in the sections were 15.18, 28.06, 48.76, and 77.22 w/o NpO_2 , with the balance of material being calcia-stabilized zirconia. Conventional ball-milling, cold-pressing, and sintering techniques were utilized for powder blending and pellet fabrication. Chemical analysis studies performed on selected pellets showed that total gas, moisture, and hydrogen contents, were considerably less than specified tolerances. Metallographic examination revealed a uniform porosity and grain structure which indicated that the distribution of constituents was homogeneous. This fact was essentially confirmed by autoradiographic examination. Limited thermal expansion and thermal cycling tests were performed on various pellets to confirm their conformance to design specifications and resistance to fracture during irradiation. All work was conducted according to rigid pellet fabrication and weld specifications as well as stringent quality assurance procedures for each phase of the assembly operation.

INTRODUCTION

By the end of the decade the needs for Pu^{238} are expected to exceed, by appreciable margins, present capabilities for production of this isotope within the USAEC Laboratories. This projection, to a great extent, depends heavily on continuation of existing space programs, but no major realignment is foreseen that would circumvent the need for radioisotope power sources. On this basis, then, one must look to commercial reactors as the only truly economical means of meeting the required production capacity.

A theoretical study of the feasibility of converting Np^{237} to Pu^{238} in commercial water reactors⁽¹⁾ was conducted by Southern Nuclear Engineering (SNE) under sponsorship of the Isotopes and Materials Branch of the USAEC in 1969. This work concluded that the approach was practical and the cost of Pu^{238}

* Owned by Connecticut Yankee Atomic Power Company.

could be reduced appreciably with preservation of adequate profit margins for the utilities. Questions still remained regarding limitations of the one-dimensional model used for the calculations and the unavailability of experimental data related to the characteristic behavior of neptunium in a light-water-reactor neutron environment. A project definition phase of a program aimed at generation of experimental and analytical data necessary to the critical analyses of the concept through use of a one-dimensional finite difference computer model was initiated by SNE in 1970⁽²⁾. This effort led to an agreement whereby Battelle's Columbus Laboratories (BCL) was responsible for the fabrication of two NpO_2 target assemblies which were to be irradiated in Connecticut Yankee Reactor owned by Connecticut Yankee Atomic Power Company (CYAPC). Design drawings with full specifications, and the results of multidimensional calculations for the target rods in a pressurized water reactor have been reported⁽³⁾.

Specifications called for the fabrication of two target rod assemblies, each consisting of four separate sections with different neptunium compositions and mass-volume relationships, as listed in Table 1. The target material was to be composed of neptunium dioxide mixed with a suitable diluent in the form of pellets sintered to a fairly high density to promote heat transfer. Seamless Type 304 stainless steel tubing was specified as the cladding material. Fin-shaped spacers were located on the top ends of each segment of the targets to position the rod within the thimbles of the control rod assemblies which were chosen for the irradiation.

Battelle's responsibility in the overall program was focused on experimental development and conduct of full quality assurance procedures for cladding examination, pellet fabrication, pellet characterization, some physical property measurements, rod segment welding, shipment, and final assembly and welding at the reactor site. This report covers the specifications and detailed process operations that were employed to fulfill this responsibility.

TABLE 1. SPECIFIED NEPTUNIUM CONTENT OF
VARIOUS TARGET ROD SEGMENTS

Segment		Neptunium Density, g per cm ³	Total Volume of Fuel Segment, cm ³	Total Mass of Neptunium, g
1A	2A	5.44	18.88	102.69
1B	2B	2.72	18.88	51.35
1C	2C	1.36	18.88	25.67
1D	2D	0.68	81.17	55.20

MATERIAL SELECTION

Target Constituents

Only a few properties of NpO_2 have actually been reported in the literature, and to complicate the situation further there are virtually no data relating to fabrication behavior. Ackermann, et al.⁽⁴⁾, previously worked with small quantities of NpO_2 to study its vaporization characteristics. Their work showed that NpO_2 was the principal vapor species although a tendency to become hypostoichiometric and form a lower oxide (probably Np_2O_3) through loss of NpO was found at high temperatures. In this respect the phase relationships found in the neptunium-oxygen system seem to closely resemble those established for plutonium-oxygen.

The theoretical density of NpO_2 was calculated to be 11.13 g per cm^3 based on the lattice constant value of 5.434 Å reported by Ackermann, et al.⁽⁴⁾. This information was of immense value throughout the program since it provided a means for calculation of reliable theoretical densities of neptunia solid solutions with the diluent material. These data in turn aided the interpretation of the sintering behavior as the approach to theoretical density was realized. Since little more was known of the intrinsic behavior of NpO_2 an assumption was made that its melting point, and phase interactions, would closely parallel those of UO_2 and PuO_2 based on the periodic table relationships of this family of actinide compounds. Moreover, the sintering characteristics of NpO_2 were assumed to be about the same as those of UO_2 and PuO_2 , which effectively bracketed the temperature range for attaining maximum densification between 1500 and 1700 C.

The optimum target appeared to be columns of pellets consisting of a dispersion of NpO_2 in some diluent added to reduce self-shielding of the neptunium. The term dispersion, as used here, is probably a misnomer since some solubility, perhaps complete, would be predicted under the sintering conditions imposed to obtain pellets with a high degree of resistance to fracture and spalling coupled with the optimum attainable heat transfer. Materials and techniques normally used for target fabrication⁽⁵⁾, e.g., powder metallurgical preparation of aluminum compacts with a uniform dispersion of NpO_2 , were unsuitable for this study because of the high temperatures involved in the irradiation. As a means of making a selection a list of some of the more important desired

characteristics of the diluent material was formulated, as given below:

- (1) Low neutron cross section
- (2) Good irradiation stability in a water reactor neutron spectrum
- (3) Excellent high-temperature stability, e.g., to temperatures near the melting point of NpO_2 or $\sim 2000^\circ\text{C}$ (3632°F)
- (4) High probability of predictable uniform sintering characteristics based on behavior with other actinide dioxides, e.g., UO_2 and PuO_2
- (5) Compatible with Type 304 stainless steel cladding at 600°F , the normal reactor water temperature during operations.

Other factors taken into consideration were the low calculated heat evolution from the target rod pellets and almost negligible problems of swelling from fission products, etc., normally associated with reactor fuels.

Based on the above criteria a decision was made to utilize CaO-stabilized ZrO_2 in solution with the NpO_2 . ZrO_2 was considered to be more favorable than Al_2O_3 because of possible deleterious side reactions associated with aluminum. Calcia stabilization of the zirconia appeared mandatory because of the complete lack of data on the $(\text{Np},\text{Pu})\text{O}_2$ - ZrO_2 system and the wide range of compositions called for in the target specifications.

Neptunium dioxide for this study was obtained from the Atomic Energy Commission via the Savannah River Plant. The powder was light green in color and was received with the specification sheet shown in Table 2. As a general observation, the impurity content of the NpO_2 powder, as determined by spectrographic analysis, was about the same as that expected in commercial-grade UO_2 and PuO_2 powders when the limits of detection and the spectrum of elements examined were evaluated. The low plutonium content of this material was considered to be essential for the planned irradiation to avoid excess fissioning within the targets.

The calcia-stabilized zirconia powder for use as the diluent with the neptunium dioxide was procured from the Zirconium Corporation of America (ZIRCOA). This material was ordered as nuclear-grade powder containing 10 to 12 w/o calcia of minus 325 mesh ($< 44\text{-}\mu$ particle size). A typical chemical analysis for impurities in the powder furnished by ZIRCOA is given in Table 3 together with the results obtained at Battelle-Columbus. As can be seen, the Battelle analysis is in fairly good agreement with that of the manufacturer. The relatively high silicon and sulfate contents of the powder were recognized as factors which could lead to fabrication difficulties but no other substitute material could

TABLE 2. RESULTS OF CHEMICAL ANALYSES ^(a) ON NpO₂
POWDER RECEIVED FROM SAVANNAH RIVER

Run Identification	010HB-14
Oxide Weight, g	850.0
Neptunium Oxide Assay, %	99.54
Neptunium in Neptunium Oxide, %	88.1
Neptunium Weight, g	745.37
Thorium, w/o oxide	0.14
Uranium, w/o oxide	L 0.10
Weight loss at 800 C, w/o oxide	0.21
Total Plutonium w/o oxide	0.02

Plutonium Isotopic Distribution

²³⁸ Pu	78.32
²³⁹ Pu	18.98
²⁴⁰ Pu	2.19
²⁴¹ Pu	0.376
²⁴² Pu	0.140

Spectrographic Analysis, ppm

Al, 150	Fe, 200	Ni, L50
B, L20	K, L 200	P, L500
Be, L10	Mg, 10	Pb, L10
Ca, L50	Mn, L 50	Si, L100
Cd, L10	Mo, L 10	Sn, L50
Cr, 30	Na, L 20	Zn, L200
Cu, L50		

(a) L = less than.

TABLE 3. TYPICAL AND ANALYZED IMPURITIES IN THE
CALCIA-STABILIZED ZIRCONIA POWDER^(a)

Impurity	Impurity Content, ppm	
	Typical ZIRCOA Analysis	BCL Analysis
Aluminum	100-300	40
Boron	0.5-2	<5
Cadmium	<10	<10
Chlorine	10-20	40
Cobalt	<10	<10
Chromium	10-30	10
Copper	<25	<10
Iron	85-400	150
Hafnium	50-100	<100
Magnesium	100-150	50
Manganese	<10	5
Nickel	N.A.	20
Lead	<25	<10
Silicon	280-600	300
Tin	≈10	<10
Titanium	<10	20
Vanadium	≈10	<10
SO ₄	100-400	30
Loss on ignition	0.3% max	N.A.

(a) ZIRCOA reactor-grade zirconia containing 10 to 12 w/o CaO.
N.A. = not analyzed.

be obtained in time to meet the commitments of the program. Removal of chlorine during sintering was expected; therefore, the high chlorine content of the powder did not present any cause for concern.

The stabilized zirconia powder was then examined by metallographic techniques in an effort to further characterize the material before blending with the neptunia. A photomicrograph from the mounted and polished powder specimen is shown in Figure 1. As expected, the particles were irregular in shape, and a cursory size analysis indicated that approximately 80 percent of the powder was $\leq 10 \mu$ in maximum width. No similar evaluation of the NpO_2 was conducted; however, it was concluded that the particle size of the two constituents was sufficiently fine to give a highly sinterable powder blend with reasonable ball-milling periods.

Cladding Components

Type 304 stainless steel tubing for the target rods was supplied by Nuclear Materials and Equipment Corporation. This tubing, which conformed to the specification NPS-20A-Rev 1 (Reference NUMEC Drawing 10B323) was completely qualified for use in Connecticut Yankee core reload assemblies. The four tubes received for this program were each approximately 10 ft long, and these were identified as S-557, -562, -566, and -569. Dimensions of this reactor-grade fuel rod tubing were 0.433 in. in OD with a 0.0165-in. wall thickness. Results of chemical analyses for each of the tubes are presented in Table 4, along with the mill heat analysis and ASTM and SNE specifications. In general, all of the elements were within the standard specifications with the exception of the chromium content of Tube S-569, which was slightly on the low side. Battelle analyses for gaseous impurities fall within the ranges normally expected for these elements.

Further characterization included metallographic examination of transverse and longitudinal sections from each of the tubes. Typical microstructures of the various tube sections are shown in the photomicrographs of Figures 2 and 3 for Tube S-566. All of the sections were examined first in the as-polished condition to obtain a true picture of the inclusion distribution and then in the etched condition for grain size evaluation. Small amounts of stringer-type inclusions seen in Figure 2b were present in all of the specimens examined in the longitudinal direction. These stringers were often found close to the

500X

As Polished

2F350

FIGURE 1. TYPICAL CaO -STABILIZED ZrO_2 PARTICLE
CONFIGURATION AND SIZE DISTRIBUTION

TABLE 4. CHEMICAL ANALYSIS DATA FOR TYPE 304 STAINLESS STEEL TARGET TUBE MATERIALS^(a)

Element	Specification Analysis		Mill Analysis ^(d)		Battelle Analysis of Indicated Tube ^(e)			
	NUMEC ^(b)	SNE ^(c)	Heat	Check	S-557	S-562	S-566	S-569
Oxygen	-	-	-	-	150 ppm	200 ppm	175 ppm	235 ppm
Hydrogen	-	-	-	-	4 ppm	7 ppm	4 ppm	3 ppm
Nitrogen	-	-	-	-	510 ppm	660 ppm	640 ppm	520 ppm
Carbon	0.08 max	0.04-0.08	0.04	0.04/0.04	0.038	0.041	0.037	0.035
Phosphorus	0.040 max	-	0.025	0.008/0.008	0.026	0.027	0.025	0.024
Sulfur	0.030 max	-	0.015	0.010/0.009	0.013	0.013	0.012	0.013
Chromium	18.0-20.0	-	18.89	18.30/18.30	19.6	18.7	18.5	17.8
Nickel	8.00-11.0	-	10.17	9.94/9.88	10.2	10.5	9.9	9.8
Manganese	2.00 max	-	1.17	1.13/1.15	1.23	1.01	1.13	0.94
Silicon	0.75 max	-	0.32	0.343/0.347	0.28	0.28	0.41	0.3
Cobalt	-	0.08 max	0.028	0.002/0.002	0.070	0.070	0.075	0.075
Boron	-	-	0.0005	<0.0005/<0.0005	N.A.	N.A.	N.A.	N.A.

(a) Analysis data given as w/o unless otherwise indicated.

(b) NUMEC specification indicates material to conform to ASTM Standard A-213-66.

(c) SNE specification is same as NUMEC except for carbon and cobalt, as indicated.

(d) Mill analysis provided by Superior Tube Company for Heat No. M2918. Check analysis is at finish of processing.

(e) Samples taken from sections removed from one end of each tube. N.A. = not analyzed.

100X

As Polished

2F340

a. Transverse Section

100X

As Polished

2F341

b. Longitudinal Section

FIGURE 2. UNIDENTIFIED INCLUSIONS (DARK) IN MATRIX OF SEAMLESS
TYPE 304 STAINLESS STEEL TUBING

100X

Etched

2F342

a. Transverse Section

100X

Etched

2F343

b. Longitudinal Section

FIGURE 3. TYPICAL MICROSTRUCTURE OF TYPE 304 STAINLESS STEEL TUBING SHOWING GRAIN STRUCTURE AND STRINGER INCLUSIONS

surface, as is evident in Figures 2b and 3b. Generally, the microstructures were typical of those expected for this grade of stainless steel tubing. One of the tubes, S-566, appeared to have a somewhat higher concentration of inclusions; therefore, this length of tubing was held in reserve and utilized for preliminary testing of weld parameters and as cladding for compatibility capsules. Boroscope examination was conducted on all tubing after cutting to size. Some small pits and scratches were identified on the interior of all of the tubes but in no case were these considered to be of sufficient magnitude to preclude the use of a tube for a target segment. However, as a result of this evaluation all tubes were classified and subsequently utilized according to this listing.

Type 304 stainless steel rod for the various end fittings and other hardware components was procured in accordance with the SNE specification (Reference Drawing SNE69-646-11). The Battelle chemical analysis of this material is given in Table 5 together with the SNE specifications and the certified mill analysis. Concentrations of all elements were within the expected ranges. The high cobalt content allowed for the stainless steel rod was appreciably above the generally accepted limit; however, it was necessary to relax the specification for cobalt somewhat in order to obtain material that would satisfy the immediate needs of the program. This bar stock was the only lot of stainless steel commercially available at the time the order was placed. Although stainless steel with a lower cobalt content can be obtained on special order, several months are required for delivery and the quantity of steel ordered must be appreciably greater than warranted for this investigation.

Cursory evaluation of the effects associated with the high cobalt content of the cladding end fittings indicated that some degree of perturbation of the gamma scans of the target rods would be expected on postirradiation examination, but no serious or insurmountable problem was thought to exist. Metallographic examination of transverse and longitudinal center sections from the rod stock yielded the expected microstructures, as shown in Figure 4. Comparison of these microstructures with those of Figures 2 and 3 shows that the average grain size of this material was considerably greater than that of the cladding.

TABLE 5. CHEMICAL ANALYSIS OF TYPE 304 STAINLESS STEEL ROD
USED FOR TARGET END COMPONENTS AND OTHER HARDWARE^(a)

Element	Specification Analysis ^(b)	Certification Analysis ^(c)	Battelle Analysis
Oxygen	-	-	600 ppm
Hydrogen	-	-	< 1 ppm
Nitrogen	-	-	300 ppm
Carbon	0.04-0.08	0.057/0.059	0.065
Phosphorus	0.045 max	0.022/0.023	0.027
Sulfur	0.030 max	0.021/0.019	0.025
Chromium	18.00-20.00	18.40/18.50	18.5
Nickel	8.00-12.00	8.36/8.36	8.25
Manganese	2.00 max	1.51/1.52	1.65
Silicon	1.00 max	0.52/0.52	0.46
Cobalt	0.15 max	0.13/0.13	0.12
Molybdenum	-	0.26/0.26	N.A.
Copper	-	0.20/0.20	N.A.

(a) Analysis data in w/o unless otherwise specified. N. A. = not analyzed.

(b) SNE Drawing 69-646-11 specifies ASTM Standard A-276, Type 304, centerless ground, Condition A, except for carbon, 0.04 to 0.08 w/o, and cobalt, 0.15 w/o max.

(c) Certified analysis from Williams and Company on Universal Cyclops Heat No. G5433.

100X

Etched

2F381

a. Transverse Center Section

100X

Etched

2F383

b. Longitudinal Center Section

FIGURE 4 MICROSTRUCTURE OF TYPE 304 STAINLESS STEEL ROD STOCK

PELLET PREPARATION

Target Specifications

Initially it was thought that extensive parametric processing studies could be avoided since normal stringent performance characteristics associated with a reactor fuel were not specified for the targets and there was no necessity for maintenance of excessively precise restrictions on density. However, as the work progressed and the goals of the program became more well defined it became obvious that there was a need for target pellet specifications equivalent in many ways to those for the fuel so as not to jeopardize the operation of the reactor. This was further deemed necessary when the additional value of experimental data generated on pellets with a uniform density was considered in light of more ambitious goals for the program. In addition, a rigid diametral tolerance gave some assurance that appreciable void volume would form within the targets if the fuel-cladding gap closed during the course of the irradiation. The requirement for uniform clearance thus minimized the probability of breachment of the cladding. Control of pellet diameter was also found to be of significance since this dimension affects the NpO_2 concentration per unit length, which when held constant provides a means of mapping the flux of the reactor through determination of the change in Pu^{238} concentration over the length of the target segment.

In view of the above, a pellet specification was written toward the completion of the program. This document is given as the Appendix to this report. The standards imposed on the target material closely parallel those normally required for UO_2 fuel. The only exceptions occur in the allowable impurity elements, particularly in regard to the total equivalent boron content since reactivity of the target rod was of no consequence, and in the dimensional limits of surface flaws. Otherwise, the acceptable amount of chlorine, hydrogen, and total gas was the same as for the reactor fuel pellets. These limits and the consequences of preparing pellets that could not meet the specifications were factors that profoundly influenced the techniques chosen for pellet fabrication; these are discussed below.

Target Fabrication

Since data pertaining to the densification behavior of NpO_2 were virtually nonexistent, initial studies were directed toward obtaining a cursory understanding of the powder blending characteristics and the change in density that could be expected on sintering the various compositions of interest. It was expected that this work would then provide the basic information needed to make adjustments in the processing scheme and thus zero in on the fabrication parameters that would meet the goals established for this phase of the program.

Neptunium concentrations specified for each of the target segments are given in the tabulation of Paragraph 5.1 of the Appendix. These values, which range from 77.22 to 15.18 w/o NpO_2 , essentially encompass the entire phase system in the pseudobinary NpO_2 -(10 w/o CaO-ZrO_2). Accordingly, an appreciable change in blending and densification characteristics could be expected for these compositions due to the tremendous difference in properties between the components.

As a first step in the investigation, various techniques for blending of the NpO_2 and 10 w/o CaO-ZrO_2 powders were evaluated according to the relative probability of obtaining a homogeneous dispersion of NpO_2 in the sintered pellets. This was necessary because many standard methods are unsuitable for blending of powders with widely different densities, as is the case with NpO_2 and calcia-stabilized ZrO_2 , which have densities of 11.13 and about 5.0 g per cm^3 , respectively. A decision was eventually made to blend the powders by wet ball milling, on the basis of experience at Battelle with mechanically mixed UO_2 and PuO_2 together with an evaluation of previous work on UO_2 - PuO_2 - ZrO_2 fuels⁽⁶⁾.

Approximately 30-g powder batches* of various compositions were blended for the preliminary sintering trials. Each powder batch was blended for 8 hr in a rubber-lined mill containing zirconia balls and filled with distilled water. The powder slurry was then removed, dried, and screened prior to

* As defined in the Appendix, Paragraph 7.2.1, a batch of powder is defined as a blend of the initial constituents to obtain a specific composition.

pressing. Considerable foaming occurred during milling due to the sulfate content of the stabilized zirconia powder, but aside from some inconvenience on cleaning there was no deleterious effect on the dried powder. All work on this and other phases of the effort connected with pellet fabrication was conducted in glove boxes containing a dry nitrogen atmosphere. Pellets approximately 0.45 in. in diameter and 0.65 in. long were uniaxially cold pressed at 20,000 and 40,000 psi and sintered in a high-temperature resistance furnace employing a tungsten mesh heater at temperatures ranging from 1300 to 1800 C. An atmosphere of either flowing argon-8 v/o hydrogen or pure nitrogen was used throughout the sintering cycle. Pellets were placed on one of several tiers of a tungsten holder, which was positioned well within the hot zone of the furnace heater. Densities of the pellets were determined from their physical dimensions and weights. Theoretical densities of the various target segment compositions were calculated from a derived theoretical density for NpO_2 , based on the lattice constant data, together with an estimated density for 10 w/o CaO-ZrO_2 obtained from the manufacturer.

A compilation of the various process parameters investigated and the resulting densities of the pellets is given in Table 6. Duplicate pellets of the 48.76 and 77.22 w/o NpO_2 compositions were cycled to progressively increasing temperatures to determine the effect of temperature on shrinkage behavior. In addition, small amounts of mechanically dry-blended powder containing 77.22 w/o NpO_2 , together with the as-received NpO_2 , were pelletized and sintered at increasing temperatures to obtain a measure of the benefits of reduction in particle size through ball milling. As soon as the data indicated that densities on the order of 90 percent of theoretical could be attained by high-temperature firing, batches of several hundred grams of powder were wet blended for a period of 24 hr and subsequently evaluated for densification characteristics.

Generally, sintering temperatures on the order of 1700 or 1800 C were needed to attain densities near 90 percent of theoretical with powder that was wet milled for 8 hr. Shrinkages were quite high and comparatively nonuniform, i.e., shrinkage was far greater in the radial direction than in the longitudinal direction and pellets were hourglass shaped or tended to be larger in diameter at one end. This behavior was attributed somewhat to the pressing techniques, but powder characteristics were thought to be primarily responsible. This was evident from the number of fissures, blow holes, and in some cases raised areas

TABLE 6. RESULTS OF INITIAL SINTERING STUDIES OF VARIOUS NpO_2 TARGET ROD PELLETS

Segment	Nominal Composition, w/o NpO_2	Ball-Milling Time, hr	Densification Pressure, psi	Sintering Temperature, C	Sintering Atmosphere	Density of Material		Diametral Shrinkage, percent
						G per cm^3	Percent of Theoretical ^(a)	
D	15.18	8	40,000	1700	Ar-8 v/o H_2	4.93	90.2	16
		8	40,000	1700	Ar-8 v/o H_2	4.98	91	-
		8	20,000	1700	Nitrogen	4.92	90.2	17
		8	20,000	1800	Nitrogen	5.07	92.7	17.6
		24	20,000	1800	Nitrogen	5.22	95.5	13.2
		24	20,000	1800	Nitrogen	5.25	96	13.3
		24	20,000	1700	Nitrogen	5.18	94.8	12.7
		24	20,000	1600	Nitrogen	4.97	91	11.4
		24	20,000	1675	Nitrogen	5.17	94.6	-
		24	20,000	1675	Nitrogen	5.16	94.4	12.4
		24	20,000	1675	Nitrogen	5.13	93.8	12.7
C	28.06	8	40,000	1700	Ar-8 v/o H_2	5.37	90.7	-
		8	40,000	1700	Ar-8 v/o H_2	5.28	89.1	-
		8	20,000	1700	Nitrogen	5.38	91	18.2
		8	20,000	1800	Nitrogen	5.38	91	18.1
		24	20,000	1675	Nitrogen	5.47	92.4	-
		24	20,000	1675	Nitrogen	5.39	91	12.9
B	48.76	8	20,000	1300	Nitrogen	4.53	66.3	9.1
		8	20,000	1500	Nitrogen	5.13	75	13.3
		8	20,000	1700	Nitrogen	6.04	88.3	17.2
		8	20,000	1800	Nitrogen	6.03	88.3	17.6
		24	20,000	1675	Nitrogen	6.37	93.2	13.7
A	77.22	8	20,000	1300	Nitrogen	5.67	65.2	11.1
		8	20,000	1500	Nitrogen	7.16	82.5	17.6
		8	20,000	1700	Nitrogen	7.15	82.5	18.0
		8	20,000	1800	Nitrogen	7.74	89	19.9
		24	20,000	1675	Nitrogen	8.70	100	16.8
		24	20,000	1675	Nitrogen	8.54	98.2	16.5
A	77.22	Dry Blended	20,000	1400	Nitrogen	5.63	64.7	9.3
				1500	Nitrogen	6.04	69.4	11.4
				1600	Nitrogen	6.58	75.6	14.2
				1700	Nitrogen	7.14	82	16.9
				1800	Nitrogen	7.14	82	-
-	100	As Received	20,000	1400	Nitrogen	8.55	76.6	-
				1500	Nitrogen	8.69	78	-
				1600	Nitrogen	-	-	-
				1700	Nitrogen	8.71	78	-
				1800	Nitrogen	8.71	78	-

(a) Based on calculated theoretical densities of the 15.18, 28.06, 48.76, 77.22 and 100 w/o NpO_2 compositions of 5.46, 5.92, 6.84, 8.70, and 11.13 g per cm^3 , respectively. Calcia-stabilized ZrO_2 theoretical density is estimated to be 5.0 g per cm^3 .

(b) Same pellet(s) sintered at increasing temperatures.

seen on the surface of the pellets which were apparently caused by impurities present in the as-received powders.

In comparison, pellets pressed from the dry-blended and as-received powders attained densities of only 82 and 78 percent of theoretical when sintered at temperatures up to 1800 C. This behavior demonstrated the greater activation of the wet-milled powder and enhanced densification through sintering that could be obtained by particle reduction. An additional and significant observation was the similarity among the surface characteristics of the pellets prepared from the dry-blended, as-received, and wet-milled powders. Further examination of the pellets by standard metallographic techniques showed that the porosity distribution was for the most part quite uniform except for occasional large void areas which appeared to be caused by gas formation from impurities. These voids were generally of less magnitude in the pellets prepared with wet-milled powders. This information together with the somewhat greater dimensional uniformity of these pellets suggested that a longer ball-milling time would be of value in enhancing ultimate densification and reducing surface defects. No evidence of metallic impurities was found in any of the microstructures and all specimens appeared to have a fairly equiaxed grain configuration indicative of a single-phase $\text{NpO}_2\text{-CaO-ZrO}_2$ solid solution. Similarly, there was no indication of the presence of the ZrO_2 monoclinic phase when the specimens were examined with polarized light.

Further reference to Table 6 shows that there was no increase in sintered density from either the use of high densification pressure, e.g., 40,000 psi as compared to 20,000 psi, or the change of sintering atmosphere from pure nitrogen to argon-8 v/o hydrogen. Data pertaining to the densification characteristics of pellets prepared from the large batches of powder milled for 24 hr are also shown. A general increase in density was noted with the longer ball-milling time for all compositions but the change was more significant in the pellets with the higher NpO_2 content. This trend is more readily seen by comparison of the data pertaining to the 77.22 w/o NpO_2 powder batches where material ball milled for 8 hr achieved a density of only 82.5 percent of theoretical on sintering to 1700 C, while the 24-hr-milled powder treated in the same manner sintered to densities close to the calculated theoretical value. Another quite noticeable change occurred in the shrinkage characteristics of the powder milled for the longer period. By referral to Table 6 it can be readily seen that the diametral shrinkage of pellets with

sintered densities in the range of 90 to 95 percent of theoretical was considerably less than found in earlier work. For example, initial shrinkage values generally fell within the range of 17 to 18 percent when the various powder blends were sintered at about 1700 C for 2 hr. Calculations also indicated that the shrinkage was nonuniform, with the ratio of the diametral to longitudinal change being about 1.7. In comparison, the somewhat finer powder produced by milling for 24 hr generally showed a diametral change of about 12 percent and more uniform shrinkage behavior, with a ratio of the percent change in diameter to length of about 1.1. The trend was similar to that observed for overall densification, i.e., the greatest amount of diametral shrinkage occurred in the 77.22 w/o NpO_2 composition and the dimensional nonuniformity or degree of hourglass configuration was also greatest in this material.

A cursory study of the sintering characteristics of the 24-hr-milled 15.18 w/o NpO_2 powder showed that hourglassing could be minimized by sintering at 1800 C; however, the densities of these pellets were near 96 percent of theoretical, which was greater than desired since by this time all process parameters were adjusted to the projected 90 percent density value. When the sintering temperature was lowered to 1600 C, dimensions of the pellets were generally somewhat irregular, indicative of nonuniform shrinkage within the fired specimens. Although the calculated densities of pellets sintered at 1600 C were very close to the desired value of 90 percent of theoretical the best compromise in regard to dimensional stability and appearance was obtained by treatment at 1675 C. Time did not permit the same type of study with other compositions. In any event, the 1675 C sintering temperature was found to be satisfactory for the 28.06 and 48.76 w/o NpO_2 compositions as well. A decision was made to lower the sintering temperature of the 77.22 w/o NpO_2 pellets to 1600 C on consideration of the near theoretical densities attained by firing at the slightly higher temperature of 1675 C.

Throughout the above effort, shrinkages were obtained on one or two pellets of a given composition and these data were then used to calculate the die sizes needed to prepare densified pellets within several thousandths of an inch of the specified mean diameter of 0.3825 in. Early results immediately indicated the difficulties associated with an original intent to sinter the pellets to size. Therefore, dry centerless grinding was incorporated into the fabrication process as an additional but necessary step to insure uniform clearance between the pellets and cladding wall.

A flow diagram outlining the various process steps utilized in the fabrication of the target pellets is shown in Figure 5. This general scheme was used to prepare six batches* of pellets, as described in Table 7. All pellets were centerless ground to a diameter of 0.383 ± 0.001 in. The densities shown in Table 7 were obtained on randomly selected pellets. All values were quite close to the mean for the various batches of material. This excellent reproducibility was achieved only through exceedingly close control of all process variables including pressing techniques, heating and cooling rates during sintering, and gas flow through the furnace.

Zirconia Pellet Fabrication

According to specifications, each end of the four target segments composing the two rods required nonfissile zirconia pellets to act as barriers between the hot center of the target columns and the stainless steel components. Characterized calcia-zirconia powder, obtained for preparation of the target pellets, was also used for this purpose.

Pellets were pressed from the stabilized zirconia powder at a pressure of 60,000 psi and subsequently sintered at 1800 C for 2 hr. Heating and cooling rates ranged between 200 and 300 C per hr. A flow of pure nitrogen was maintained through the furnace during the entire sintering cycle.

Dimensions of the densified pellets were 0.433 to 0.438 in. in diameter by about 0.5 in. long. All pellets were dry centerless ground to a diameter of 0.384 ± 0.001 in. and ends were faced to leave the pellets at their maximum length. Determinations of the density of several pellets gave values ranging between 90 and 93 percent of theoretical.

* Paragraph 7.2.1 of the Appendix defines a pellet batch as those compacts pressed and sintered under identical conditions in the same furnace cycle.

FIGURE 5. PROCESS FLOW DIAGRAM FOR PELLET FABRICATION

TABLE 7. PROCESS PARAMETERS AND DENSITIES OF NpO_2 TARGET PELLETS

Target Segment	Batch Identification	Nominal Composition, w/o NpO_2	Densification Pressure, psi	Temperature of Nitrogen Sinter, C	Density, ³ g per cm^3	Density Percent of Theoretical
1A	4	77.22	20,000	1600	8.61	99.0
					8.65	99.4
					8.65	99.4
2A	4	77.22	20,000	1600	8.61	99.0
					8.61	99.0
					8.61	99.0
1B	5	48.76	20,000	1675	6.15	90.0
					6.13	89.7
					6.16	90.0
2B	5	48.76	20,000	1675	6.18	90.4
					6.14	90.0
					6.20	90.6
1C	6	28.06	20,000	1675	5.42	91.6
					5.42	91.6
					5.51	93.0
2C	6	28.06	20,000	1675	5.31	89.7
					5.45	92.1
					5.40	91.3
1D	2	15.18	20,000	1675	5.10	94.0
					5.11	94.0
					5.13	94.6
	1	15.18	20,000	1675	5.12	94.5
					5.13	94.6
					5.08	93.6
2D	3	15.18	20,000	1675	5.03	92.8
					5.04	92.9
					5.12	94.5

PELLET CHARACTERIZATION

Chemistry

Two pellets were selected from each batch of material for chemistry studies. One of these was used for immediate analysis of total gas, moisture, spectrographic impurities, NpO_2 , CaO , and ZrO_2 . The other pellet was stored as an archive sample to be available if needed during examination of the targets after irradiation. Specifications also called for the determination of the typical hydrogen content of the gas released from the pellets and measurement of the chlorine content of a typical 15.18 w/o NpO_2 pellet. The latter analyses were to serve as backup data if total gas and moisture approached or exceeded specifications.

Gas collection and moisture analysis were conducted in low-volume glass apparatus that had been thoroughly outgassed by heating and evacuation. The apparatus was divided into two sections by appropriate valves, with the lower section containing the pellet and moisture probe and the upper section, a pressure gage and collection bulb. The analytical procedure consisted of heating a pellet to 1000 C in a system previously evacuated to 10^{-6} torr with a mercury diffusion pump, followed by immediate collection and analysis of all evolved gas. Each pellet was held at temperature for 30 min. The moisture content of the gas was monitored continually by an EMF probe^{*}, and the amount of total gas was obtained by measurement of the pressure changes in the calibrated volume portion of the apparatus.

The results of these analyses are given in Table 8. In all cases the total gas was far less than the 0.05 cm^3 per g specified for the target pellets, and the moisture content was exceedingly low at less than 1 ppm. Although samples of evolved gas were taken from all pellets only one was analyzed, as per previous agreement with CYAPC and SNE. This approach seemed quite reasonable in view of the fact that all pellets received the same prior treatment and the total gas and moisture contents were generally very low. The values obtained from the mass spectrometric measurement of the gas from a 77.22 w/o NpO_2 pellet are given in Table 9. As can be seen, the off gas was composed

*Panametrics hydrometer, Model 1000, Waltham, Massachusetts.

TABLE 8. TOTAL GAS RELEASE AND MOISTURE CONTENTS
OF SELECTED NpO_2 PELLETS

Sample Designation	Nominal Composition w/o NpO_2	Gas Volume, cm^3 per g (STP)	H_2O , ppm
22	15 (Batch 1)	0.0022	< 0.38
36	15 (Batch 1)	0.0079	0.08
34	15 (Batch 2)	0.0050	< 0.10
38	15 (Batch 3)	0.0280	< 0.10
28	28	0.0270	0.05
30	48	0.0240	< 0.10
21	77	0.0150	0.10

TABLE 9 RESULTS OF MASS SPECTROMETRIC ANALYSIS
OF GAS FROM 77 w/o NpO_2 PELLET

Species	Amount, v/o
CO_2	0.98
Air	0.60
Oxygen	0.32
CO	57.6
Nitrogen	25.6
Hydrogen	10.1
C_2H_6	0.59
CH_4	0.16
SO_2	<0.02
H_2O	3.9
Unknown component	0.27

primarily of nitrogen and carbon monoxide. Hydrogen, which was to be held to < 10 ppm, was present in an insignificant amount.

By analogy in processing, it can be assumed that the distribution of the various species present in the evolved gas did not change appreciably with composition. It is interesting to note that conversion of the percentage moisture in the gases to ppm by weight yielded values ranging between 0.9 and 0.07, which is in relatively good agreement with the results obtained by use of the electrolytic probe. This correlation essentially substantiated earlier predictions that very little hydrogen and moisture would be found in the pellets. In addition, since the total amounts of evolved gas were all quite low, one can be quite confident that there was no introduction of impurities into a particular pellet batch that would have gone undetected by the procedures used in these analyses.

A separate determination of the chlorine content of the 15.18 w/o NpO_2 composition was conducted to obtain some indication of the amount of this impurity present after processing. A pellet of the high- ZrO_2 composition was chosen for analysis because of the high chlorine content of the as-received calcia-stabilized zirconia powder. The analytical technique employed for the measurement involved pyrohydrolysis by passage of a moisture-saturated argon gas stream over a crushed pellet while it was being heated at 950 C. The gas was passed through a solution of potassium acetate and acetic acid which was treated with mercuric thiocyanate in ethanol containing ferric alum, and this solution was then read spectrophotometrically. A value of about 8 ppm chlorine was obtained by use of this analytical method. This result showed that chlorine was removed in processing and was within the limit of 10 ppm established in the target specification. Although other batches of pellets were not analyzed for chlorine, from all indications these compositions would contain far less of this impurity than the 15.18 w/o NpO_2 material.

After completion of the total gas analysis, the pellets were broken and divided to obtain sections for general spectrographic analysis and metallography. Sections for spectrographic analysis were specifically taken from the surface of the pellets in an effort to obtain analyses that would be representative of the highest levels of impurities that could be expected in these materials. As seen from the results of this work, shown in Table 10, the impurities present in various material batches were of little consequence aside from those of iron, magnesium, manganese, chromium, and silicon. Generally acceptable impurity levels

TABLE 10. IMPURITIES PRESENT IN SELECTED PELLETS FROM VARIOUS BATCHES OF MATERIAL AFTER SINTERING

Element	Impurity Content, ppm							UO ₂
	Sample 22, 15 w/o NpO ₂ (Batch 1) ²	Sample 36, 15 w/o NpO ₂ (Batch 1) ²	Sample 34, 15 w/o NpO ₂ (Batch 2) ²	Sample 38, 15 w/o NpO ₂ (Batch 3) ²	Sample 28, 28 w/o NpO ₂	Sample 30, 48 w/o NpO ₂	Sample 21 77 w/o NpO ₂	
Bi	<2	<2	<2	<2	<2	<2	<2	2
B	<0.3	<0.3	<0.3	<0.3	<0.3	<0.3	<0.3	1
Cd	<5	<5	<5	<5	<5	<5	<5	1.0
Cr	80	95	95	95	85	80	80	500.0
Cu	11	3	5	11	3	2	2	25.0
Fe	375	280	520 ^(a)	700 ^(a)	375	125	220	500.0
Li	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	0.1
Mg	200	200	225	200	200	200	150	50.0
Mn	75	95	50	55	75	12	75	10.0
Mo	<6	<6	<11	<11	<6	<11	<6	150.0
Ni	8	<8	25	35	8	<12	<8	300.0
P	<210	<210	<210	<210	<210	<210	<210	-
Pb	40	40	<4	<4	40	<4	<40	20.0
K	<125	<125	<250	<250	<125	<250	<125	-
Si	110	70	300	300	60	80	<34	500.0
Ag	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	0.3
Na	80	50	<40	<40	80	<40	160	-
Sn	20	20	10	20	20	10	20	5.0
V	<11	<11	<11	<11	<11	<11	<11	1.0
Zn	<50	<50	<50	<50	<50	<50	<50	20.0

(a) Contaminated with iron during sampling

for UO_2 pellets are given in this table for comparison.

Chemical analysis for neptunium content was conducted according to the following procedures. Duplicate samples of the pellets weighing about 0.3 g were initially dissolved in nitric acid with slight additions of hydrofluoric acid to aid dissolution. These solutions were evaporated twice and the agglomerate was redissolved in dilute sulfuric acid. The resulting solution was then fumed to remove traces of nitric acid and passed over a Jones reductor (zinc mercury amalgam) to adjust the valence of the neptunium. Final titration was conducted with ceric ammonium sulfate.

The procedures utilized for the ZrO_2 and CaO determinations were adaptations of standard methods for these elements. Portions of the pellet were crushed and dissolved in HNO_3 -HF. Neptunium (IV) was then separated from the calcium and zirconium by absorption on a Dowex 1x8 resin. Effluent from the column containing both calcium and zirconium was then treated separately to obtain constituent analyses.

For zirconium a solution of diammonium phosphate was added to precipitate ZrP_2O_7 . This precipitate was filtered from the solution, ignited at 1000 C to form ZrO_2 , and weighed to obtain the amount present. The remaining solution was then treated with oxalic acid to precipitate calcium oxalate, which was filtered and redissolved in dilute sulfuric acid. This solution was then titrated with a standard KMnO_4 solution to obtain the amount of calcium present.

All of the wet chemical analysis results are given in Table 11. These data, for all practical purposes, are adequate to verify the nominal compositions of the initial powder blends within about 10 percent. However, there was an obvious discrepancy between nominal and measured compositions which increased with increasing neptunium content. Due to this disparity the neptunium analysis of the 77.22 w/o NpO_2 sample was repeated with more care applied toward the various steps in the procedure. Reproducibility on the second run essentially verified the first results. In backup to this effort, a follow on investigation of chemical behavior was initiated as part of the experimental program to be carried out during the next fiscal period.

Several explanations of the observed results seem reasonable but all are associated with a bias in the neptunium determination and any interpretation of the data along these lines must also take into account possible slight material gains which could have occurred during ball milling. For example, all samples

TABLE 11 SUMMARY OF MAJOR OXIDE CONSTITUENT ANALYSES
FOR VARIOUS BATCHES OF TARGET PELLETS

Nominal Composition, w/o NpO_2	Batch	Sample	NpO_2 Content, w/o	ZrO_2 Content, w/o	CaO Content, w/o	Total Constituents ^(a) , w/o
15.18	1	22 ₁	15.20	71.65	13.34	100.19 high
		22 ₂	14.35	70.64	12.93	97.92 low
15.18	1	36 ₁	14.94	71.28	11.88	98.52 high
		36 ₂	14.94	68.46	12.30	95.28 low
15.18	2	34 ₁	14.98	72.49	12.75	101.75 high
		34 ₂	14.98	74.02	12.65	100.12 low
15.18	3	38 ₁	14.91	71.55	11.79	100.21 high
		38 ₂	14.90	73.51	11.71	98.16 low
28.06	6	28 ₁	25.34	62.06	9.61	100.39 high
		28 ₂	25.25	65.04	10.01	96.92 low
48.76	5	30 ₁	43.05	48.07	8.44	99.56 high
		30 ₂	42.96	47.75	8.43	99.14 low
77.22	4	21 ₁	74.69	20.48	4.69	100.05 high
		21 ₂	68.21	20.67	4.69	93.09 low
		21 ₁	69.26	-	-	-
		21 ₂	67.92	-	-	-

(a) Values listed are the totals of the high and low determinations for each oxide component.

were dissolved with difficulty and residues would probably follow some pattern in relation to the neptunia content. Then too, interfering ions could have been present during the determination or the valence state of the neptunium may not have been adjusted properly prior to titration. In any case, future studies will pay considerable attention to techniques which will obtain rapid and complete dissolution of the samples from the pellets since this was one of the difficulties that could have contributed to the apparent errors in the results. In addition controlled potential coulometry will be utilized to obtain the neptunium analysis in a manner consistent with the latest analytical methods for the actinide element in solution.

Metallographic Examination

Standard metallographic procedures were used to mount, grind, and polish samples from pellets designated for chemistry studies. An attempt was made to obtain longitudinal sections for examination but in most cases the pellets fractured into several pieces which were not truly representative of this configuration. Microstructures of the various pellet sections are shown in Figures 6 through 11. A slight chemical attack of the pore areas was noted when the samples were etched to obtain grain structure. This behavior should be taken into account when the pore areas are compared with apparent void volumes for each composition, easily derived from the density values given in Table 7. Offhand, there appears to be a good correlation between the observed porosity and the amounts thought to exist from the density calculations, i.e., the densities of 90 to 94 percent of theoretical are consistent with the microstructures. The very high densities of 99 percent of theoretical (see Table 7) determined for the 77.22 w/o NpO_2 pellets was verified by the microstructure of this material seen in Figure 11. Only a few pores present at the triple points of the grain boundaries are in evidence. This examination together with the density results also tends to confirm the nominal composition of this material as being very close to 77 w/o NpO_2 .

The grain size for various batches of pellets was quite uniform, considering the wide range of compositions prepared. Average values determined from the photomicrographs varied from 8 to 10 μ . This factor, in conjunction with the uniformity of observed porosity in all specimens, pointed to the conclusion that the distribution of the neptunia and stabilized zirconia was homogeneous.

500X

Etched

PL8736

FIGURE 6. TYPICAL MICROSTRUCTURE OF NOMINAL 15.18 w/o NpO_2
COMPOSITION AFTER SINTERING AT 1675 C FOR 2 HR²
(PELLET BATCH 1)

500X

Etched

PL8749

FIGURE 7. MICROSTRUCTURE OF TYPICAL PELLET FROM BATCH 2
CONTAINING 15.18 w/o NpO_2

500X

Etched

PL8753

FIGURE 8. MICROSTRUCTURE OF TYPICAL PELLET FROM BATCH 3
CONTAINING 15.18 w/o NpO_2

500X

Etched

PL8741

FIGURE 9. MICROSTRUCTURE OF NOMINAL 28.06 w/o NpO_2 COMPOSITION
(PELLET BATCH 6) AFTER SINTERING AT 1675 C FOR 2 HR

500X

Etched

PL8745

FIGURE 10. MICROSTRUCTURE OF NOMINAL 48.76 w/o NpO_2 COMPOSITION (PELLET BATCH 5) AFTER SINTERING AT 1675 C FOR 2 HR

500X

Etched

PL8731

FIGURE 11. TYPICAL MICROSTRUCTURE OF NOMINAL 77.22 w/o NpO_2 COMPOSITION (PELLET BATCH 4) AFTER SINTERING AT 1600 C FOR 2 HR

Note high density typified by absence of large pore volume (black areas).

Autoradiography

The procedure utilized for autoradiography was one developed for routine examination of plutonium-containing material. These checks were conducted on each of the metallographic specimens to obtain a positive record of the uniform distribution of the component materials and as a necessary criterion to comply with the pellet specifications. As can be seen in the Appendix (Paragraph 5.11), areas with neptunia concentrations of greater than 200 μ were unacceptable.

The examination technique consisted of placing the polished and cleaned surface of the metallographic samples in contact with a very fine grain photographic film* in total darkness. To prevent alpha contamination of the film a thin sheet, < 0.001 in. of polyester⁺ was placed over the specimen surface. Previous work showed that this thin layer of polyester has very little effect on alpha particle resolution. All samples were weighted throughout the time of exposure to maintain intimate contact of the polished surface with the polyester and film. Exposure periods ranged from 20 to 30 min for all compositions, with the shorter time giving the best results for the specimens containing the highest concentration of neptunia.

Negatives produced from the developed film were enlarged eight times to obtain the autoradiographic prints shown in Figures 12a through 12f. No significant dark areas or variations in contrast, which would indicate a nonuniform distribution of neptunia were found on any of these prints. In several instances very small light areas can be seen which are attributed to incomplete removal of plutonium particles[‡] from the specimen surface during cleaning. Striations on the autoradiograph of the 48.76 w/o sample (Figure 12e) are scratches made during placement or removal of the sample from the film since an almost identical exposure showed the same type of marks on the opposite end. Furthermore, the autoradiograph of the 77.22 w/o neptunia sample (Figure 12f) shows a dark curlicue emanating from an apparent imperfection on the surface which is probably a remnant from the cloth used for cleaning. The obvious sharp delineation of surface imperfections is a good indication of the resolution of

* Kodak contrast process Ortho film.

+ Melinex polyester film, Type S.

‡ Loose contamination from the glove boxes.

8X

a. 15.18 w/o NpO_2 (Batch 1)

PL8869

8X

b. 15.18 w/o NpO_2 (Batch 2)

PL8866

FIGURE 12. AUTORADIOGRAPHS OF VARIOUS METALLOGRAPHIC SPECIMENS

8X

c. 15.18 w/o NpO_2 (Batch 3)

PL8871

8X

d. 28.06 w/o NpO_2

PL8863

FIGURE 12. (Continued)

8X

PL8864

e. 48.76 w/o NpO_2

8X

PL8858

f. 77.22 w/o NpO_2

FIGURE 12. (Continued)

the technique. Therefore, these autoradiographs generally verify the metallographic observations and are further evidence of the homogeneous nature of all the pellets.

PROPERTY MEASUREMENTS

As part of the overall evaluation of the pellets an effort was made to determine the thermal expansion characteristics of the 15.18 w/o NpO_2 material and the thermal cycling behavior of all compositions. These measurements, although not intended to be thorough investigations, were considered to be the minimum backup data needed to provide some assurance that design parameters were correct and the targets would perform as expected after placement in the reactor.

Thermal Expansion

The 15.18 w/o NpO_2 composition was selected for measurement because the possibility of a phase transformation was highest in this material and deviations from design values would have the greatest effect since these pellets make up the longest length target specimen of approximately 43 in. An existing recording dilatometer was used for the measurement. This apparatus was calibrated with a high-purity alumina specimen prior to the run with the unknown. The specimen consisted of two pellets stacked to give an overall length of about 1 in. These pellets were slowly heated to 1000 C in a period of about 12 hr. Flowing high-purity nitrogen was used as a protective atmosphere throughout the time of measurement.

The thermal expansion curve depicting dilation as a function of temperature is shown in Figure 13. No discontinuities were observed in the temperature range studied. The linear thermal expansion coefficient of 8.2×10^{-6} per C (4.56×10^{-6} per F) calculated from these data was somewhat less than expected based on an assumed behavior for NpO_2 similar to that of PuO_2 and existing data for ZrO_2 . In any case, this measurement was satisfactory to show that design values were conservative for this and probably other target segments.

FIGURE 13. THERMAL EXPANSION CURVE OF THE 15.18 w/o NpO_2 MATERIAL

Thermal Cycling

Fairly early in the program it was apparent that some degree of testing would be required to verify the thermal shock resistance of the pellets under simulated reactor conditions. Compositions thought to have the least resistance were those containing the greatest amount of zirconium dioxide, a material known to have excellent insulating properties and generally poor thermal shock behavior. Neptunia was assumed to behave similarly to urania, but the lack of properties data on this material contributed to an uneasiness regarding its actual behavior in the reactor. The possibility of pellet fracture and ratcheting of the cladding prompted considerable thought directed to cycling tests. Further incentive to conduct these tests was provided by the inadvertent rapid cooling of a furnace from high temperatures during one of the initial sintering runs which resulted in fracture of an entire batch of 15.18 w/o NpO_2 pellets.

A decision was made at this time to conduct a limited series of thermal cycling experiments that would simulate cooling of the pellets during a reactor scram, the most rapid transient expected throughout the irradiation period. To obtain a series of curves approximating this condition, SNE conducted a computer analysis which determined the rates of cooling of various radial nodes for each different target composition. The results of this work are given in the SNE report describing their experimental and analytical program⁽³⁾. An attempt was then made at BCL to approximate the curves for the centerline transients. Three thermal cycles consisting of a slow heatup, a short hold, and rapid cooling were performed on the various different target pellets. Tests were performed in a low-heat-capacity furnace to facilitate cooling. In addition, the furnace was purged with helium at a high flow rate the instant that power to the furnace was turned off. This procedure gave quite rapid cooling, but even under these conditions the actual curves obtained on the pellets deviated appreciably from the calculated ones, as may be seen in Figure 14. It should be noted that the maximum temperature reached in the various tests increased with the neptunia content of the pellets. Also, all three thermal cycles were identical; therefore, the curves given in Figure 14 are a composite of the typical behavior observed for specific compositions. Pellets containing 15.18 w/o NpO_2 (Segment D) were not cyclic tested since calculations indicated that the maximum center temperature of the target segment loaded with this material would be only several hundred degrees Fahrenheit above the ambient water temperature of the reactor.

Although the cooling curves obtained on the pellets are significantly different from the calculated centerline curves, it can be rationalized that these tests were as severe as the cyclic changes actually expected for the average radial node of the targets and, hence, are satisfactory for the purpose intended. In fact, the test performed may be overly severe because cooling was initiated from temperatures considerably higher than those calculated for the midradial nodes.

All pellets were examined closely for cracks after testing and in no case was a pellet changed from its original appearance. Therefore, these studies showed that the pellets could withstand the severest transient expected during irradiation, which could only be possible in the unlikely event of a reactor scram.

FIGURE 14. COMPARISON OF CALCULATED AND MEASURED TEMPERATURE CYCLES OF VARIOUS NpO_2 TARGET COMPOSITIONS

COMPATIBILITY TESTING

Demonstration of the long-term compatibility of the pellets with the Type 304 stainless steel cladding under conditions far exceeding those expected within the reactor was required as part of the testing phase of the program. However, no definite criteria existed as to the best conditions for these tests so a temperature of 500 C (932 F) for a period of 1 month was chosen as a satisfactory goal. This decision was based primarily on the choice of a reasonably high testing temperature which would be, in effect, an accelerated test equivalent to a 1-yr treatment of the cladding at the normal reactor operating temperature of 315 C (600 F). In addition, UO_2 and PuO_2 are known to have no solid-state interaction with stainless steel at 500 C; therefore, identical behavior for the NpO_2 target pellets would demonstrate the similarity of this material with common fuels.

One pellet of each target composition was encapsulated under conditions identical to those described later for the target rods, i.e., in helium, by TIG techniques. After heat treatment the capsules were examined in the longitudinal direction for diffusion interaction between the pellets and the cladding wall. No evidence of a solid-state reaction was found on any of the capsules; however, close examination of the cladding revealed areas of grain pullout which in some instances affected 20 percent of the cladding wall. A typical microstructure of an affected area is shown in Figure 15. Disturbed areas were found in all of the capsules but there was no indication of a systematic change in the depth of penetration with pellet composition.

At this point in the program the seriousness of the situation was assessed and steps were taken to obtain as much information as possible in the short period of several weeks available before a final decision was mandatory in regard to loading of the target rods into the reactor during the Spring refueling operation. Specifically, it was decided to section all of the metallographically mounted cladding specimens in the transverse direction to obtain a separate view of the phenomenon, which might aid interpretation. One cladding specimen with the most severe grain pullout was repolished and examined on the electron microprobe. Finally, additional capsules were made up and heat treated according to the conditions listed in Table 12. In addition, several experts on stainless corrosion within Battelle were consulted as to the possible significance of the problem and rate of attack expected at the somewhat lower temperatures

250 X

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PL8685

FIGURE 15. TYPICAL APPEARANCE OF LONGITUDINAL SECTION FROM TYPE 304 STAINLESS STEEL CAPSULE WHICH CONTAINED PELLET AFTER HEAT TREATMENT AT 500 C (932 F) FOR 1 MONTH

Note grain pullout (black areas) and grain separation (black outline) near the surface of the cladding.

TABLE 12. SUMMARY OF HEAT-TREATMENT CONDITIONS
FOR THE SECOND GROUP OF TEST CAPSULES

Capsule Identification	Description	Heat Treatment	
		Temperature	Time, days
1	Blank (helium filled)	500 C (932 F)	12
2	15.18 w/o NpO_2 pellet	315 C (600 F)	10
3	Al_2O_3 grinding medium (pieces)	500 C (932 F)	12
4	NpO_2 pellet (pieces)	500 C (932 F)	12
5	CaO-stabilized ZrO_2 pellet	500 C (932 F)	9
6	UO_2 pellet	500 C (932 F)	9

(600 F) of actual reactor operation. Though no specific cause could be sited, the general consensus of opinion was that the phenomenon would not occur at 600 F even under prolonged periods of more than a year.

Examination of the cladding sections mounted in the transverse direction confirmed the fact that the pullout was localized to a number of areas on the inner surface of the cladding. Electron microprobe examination showed that there was no penetration of neptunium, calcium, or zirconium into the cladding. The specimen was also checked for other impurities that could have inadvertently been present on the surface of the pellets, e.g., aluminum, silicon, manganese, sulfur, and chlorine. None were found. Although this examination was inconclusive as to the cause of grain pullout, it unquestionably verified the absence of diffusional attack of the cladding which was the initial goal of these studies.

Further efforts were directed toward the metallographic examination of the capsules assembled and heat treated as indicated in Table 12. Since the heat treatment was substantially shorter, significant changes in the cladding were not expected. However, by this time there was a strong indication that the grain pullout was related to sensitization of the stainless steel (precipitation of chromium carbides at the grain boundaries) and somehow tied to the numerous inclusions (see Figures 3 and 4) present in certain sections of the cladding material. Based on this assumption, the type of grain structure and grain size present after heat treatment were noted and compared to those found initially as a means of obtaining some indication of a change in the stainless steel that could be related to carbide precipitation.

Pullout to the extent seen previously was not detected in any of the capsules from this group but a blank capsule containing only pure helium and another containing the UO_2 pellet exhibited very slight grain separation in some areas, forming what appeared to be the initial stages of the process. Grains in these and some of the other stainless steel capsules heat treated at 500 C (932 F) were more equiaxed and the etchant attack produced a fairly clear delineation of the grain boundaries. In comparison, the grains of the capsule containing the 15.18 w/o NpO_2 pellet remained unchanged after the heat treatment at 600 F. No attack of the cladding was seen in any of the capsules, including those containing pure NpO_2 , pure ZrO_2 (calcia stabilized), and the alumina bonding medium. Based on these results, the pellet gas analyses, and discussions with representatives from SNS (AEQ), SNE, and CYAPC, a decision was made to proceed with the irradiation of the NpO_2 target rods. Plans were also made to conduct out-of-pile compatibility tests during Fiscal Year 1972 to obtain backup

information on the long-term lower-temperature behavior of the pellets and cladding. These tests will be quite valuable in the postirradiation assessment of changes that may occur in the cladding while the rods are in the reactor.

ENCAPSULATION AND QUALITY ASSURANCE

As a necessary part of the project all work from this point forward was conducted according to written quality assurance procedures to insure proper assembly and checking of the target rod segments. This phase of the effort included construction of a welding alignment bench, determination of welding parameters through prequalification testing, weld qualification, and writeup of procedures for assembly, welding, decontamination, visual inspection, helium leak testing, and radiography. These procedures were then submitted to SNE for approval prior to actual conduct of the various operations.

Weld Qualification

Requirements for the alignment of the assembled segmented target rods were such that only a few thousandths of an inch deviation from the center axis could be tolerated for the attachment of the end plugs to the cladding tubing. In order to meet this specification a TIG welding unit was attached to an optical bench, which was bolted to a surface ground steel channel, as shown in Figure 16. With this welder design the electrode revolves around the rod, which is held stationary in alignment fixtures. The entire assembly, including the fixtures for holding the cladding tube and end fitting, was aligned within 0.0015 in. over the span of the optical bench.

Additional specifications pertaining to welding techniques were (1) less than a 0.0015-in. rise on the weld zone, and (2) positively no abrasion of the weld. These conditions, which were quite strict for normal water reactor fuel pins, were imposed to guarantee the proper water flow through the thimbles containing the target assemblies and to prevent generation of cracks or stress concentrations after welding.

Parameters for welding, i.e., the best distance of the electrode to the piece, power setting, and rotational velocity of the electrode, were established through trials with appropriately machined end fittings and shortened tube sections. Each weld was sectioned and examined by standard

FIGURE 16. VIEW OF WELDING FIXTURE AS SET UP WITH A GLOVE BOX

metallographic techniques to determine the amount of penetration obtained with the power and rotational variations. Although the optimum weld parameters were quickly established, a slight misalignment from the center axis, sometimes as much as 0.009 in. was noted on the joined pieces. As the problem became better defined a decision was made to shim the end fittings and connectors slightly to compensate for shrinkage on cooling. This technique was quite successful in bringing the overall alignment of the welded section to within a few thousandths of an inch of the center axis, which was within the straightness of the stainless steel tubing.

Three test welds were then made under identical conditions to demonstrate repeatability of the operation. In all cases metallographic examination showed the required 100 percent penetration. A typical microstructure of a weld area is shown in Figure 17. These results completed qualification of the welding process; therefore, all appropriate fittings and connectors were joined to the cladding sections while the unit was outside the glove box. The eight circumferential welds were then examined by use of the postemulsifiable-penetrant inspection technique. This method is extremely sensitive to surface-connected discontinuities since the weld area is inspected with ultraviolet light. All welds appeared to be perfect except for two which were subsequently checked with a binocular microscope and found to have very slight surface imperfections. Although these welds were readily acceptable the sensitivity of the technique was also demonstrated to the extent that there was no question of the integrity of the welds as regards microcracking and surface defects.

At the end of this phase of the work the welding apparatus, as assembled on the alignment bench, was transferred into a glove box. Modifications were then made to the box to obtain a helium atmosphere and accommodate the electrical and gas feeds for the welding equipment. A test weld was then attempted under conditions identical to those used for qualification, but in the presence of a high-purity helium atmosphere. Metallographic examination of weld area showed that the amount of penetration was identical to that obtained by welding outside the glove box.

75X

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FIGURE 17. TYPICAL MICROSTRUCTURE OF WELD ENCOMPASSING THE
TYPE 304 STAINLESS STEEL CLADDING-END FITTING INTERFACE

Encapsulation Procedures

Centerless ground pellets were wiped clean and visually examined for defects in accord with limits described in the pellet specifications. Following a thorough examination the pellets were placed on a "V" bar to obtain a stacking length measurement prior to loading. The bar used for this purpose was scribed with marks to accurately indicate the tolerances for the pellet stack lengths in each of the two target rod segment categories, i.e., 10.000 ± 0.125 and 43.000 ± 0.125 in. At this point pellets for chemistry determinations and archive storage were selected and placed in appropriately marked containers for identification. Dimensions and weights of several pellets typical of those to be loaded were measured and geometric or bulk densities were obtained from these figures. No effort was made to keep track of individual pellets throughout processing and loading; however, different batches of pellets were carefully identified as to position within the target rod segment.

Pellet stack lengths were adjusted within tolerances either by selection or grinding of the pellets. A properly dimensioned calcia-stabilized zirconia pellet was then inserted into the appropriate target rod tube and the neptunia pellets followed. Dimensions taken from the open end of the tube verified a flush fit between pellets. A second zirconia pellet was then ground to the length necessary to obtain the specified spring pressure of 1.0 and 2.5 lb for the short (10 in.) and long (43 in.) target rod segments, respectively. Spring deflections were checked under loads of 1.1 and 2.2 lb to insure conformance to specifications.

All welds were performed with a qualified operator according to procedures and parameters established earlier in the program. The glove box, which normally contained high-purity nitrogen, was switched over to helium flow and flushed thoroughly for several hours before initiation of the pellet loading and end fitting weld operations.

Extreme care was taken to keep the open end of the tube and the end fitting in the vicinity of the weld free of contamination. The end fittings were held securely but slightly shimmed to compensate for shrinkage during cooling. Visual and dimensional examination of the welds indicated that no defects were present and the total diametrical rise did not exceed 0.002 in. Deviation of the end fittings from the center axis of the target segment was less than 0.008 in. per ft. Qualification welds made before and after a series of welds were

examined metallographically and in all cases full penetration, i.e., 100 percent of the wall thickness, was observed.

Fabrication data for each target segment are summarized in Table 13. Original design specifications are also included in this table for comparison. Generally, the as-fabricated neptunium concentrations were exceedingly close to those initially specified except in case of the Segment A values. The reason for this discrepancy was pointed out earlier during the discussion of the sintering data since it relates to the very high density obtained on the 77.22 w/o NpO_2 pellets. A decision was made to shorten the stack lengths of the "A" segments by one pellet to provide sufficient archive samples since material of this composition was limited. Average pellet diameters were generally within a few ten thousandths of an inch of the values shown. This pellet diameter of 0.383+ in. fixed the pellet-cladding gap in the range between 0.003 and 0.004 in.

Cleaning and Inspection

All rod segments were decontaminated by standard techniques, i.e., cleaning with various detergent solutions followed by several rinses with ethyl alcohol. Smear counts taken after cleaning showed that no loose contamination remained on the surface. Direct counting of the surface as well as the weld areas verified the absence of contamination on the entire surface of every target segment.

A visual check was subsequently performed on the various segments in accordance with quality assurance procedures. These were found to be free of surface defects and were approved accordingly.

Following decontamination, each rod segment was immediately subjected to a helium leak check in the following manner. Several rods, or a single one if this was all that was available at the time, were loaded into a chamber which was pressurized overnight to 150 psi with helium. A thorough calibration of the leak detector with a standard leak of $7.3 \times 10^{-7} \text{ cm}^3$ per sec was initiated concurrent with this operation to assure smooth sequential movement of the segments from one step to the next. Individual rods were then rapidly moved to a chamber attached to the leak detector and evacuated. Meter readings significant of the helium present in the chamber were taken at 5 min-intervals for a period of approximately 20 min. The rate of change of the detector with the target rod present was then compared to the rate observed under identical

TABLE 13. NpO_2 TARGET SEGMENT EXPERIMENTAL AND DESIGN DATA

Segment	NpO_2 Concentration, w/o	Stack Length, in.	Average Pellet Diameter, in.	Total Target Volume, cm^3	Total Target Weight, g	Average Segment Density g per cm^3	NpO_2 Mass, g	Neptunium Mass, g		Neptunium Content per Total Target Volume, g per cm^3	
								Actual	Specified	Actual	Specified
1A	77.22	9.468	0.3835	17.92	152.62	8.52	117.85	103.83	102.69	5.79	5.44
2A	77.22	9.406	0.3835	17.80	152.33	8.56	117.63	103.64	102.69	5.82	5.44
1B	48.76	9.937	0.3835	18.81	116.14	6.18	56.63	49.89	51.35	2.65	2.72
2B	48.76	9.937	0.3837	18.83	116.84	6.20	56.97	50.19	51.35	2.67	2.72
1C	28.06	9.906	0.3834	18.74	100.53	5.36	28.21	24.85	25.67	1.33	1.36
2C	28.06	9.906	0.3838	18.78	101.00	5.38	28.34	24.97	25.67	1.33	1.36
1D	15.18	43.030	0.3834	81.41	412.85	5.07	62.67	55.21	55.20	0.68	0.68
2D	15.18	43.046	0.3835	81.48	410.90	5.04	62.37	54.95	55.20	0.67	0.68

conditions with the chamber empty and the blank rate after pumping for a period of several hours which was usually about $1.7 \times 10^{-8} \text{ cm}^3$ per sec. Analysis of the data indicated that the rate of helium dissipation found for the various rod segments was almost identical to that for the empty chamber and that readings less than the standard leak rate were attained within several minutes after the start of pumpdown.

Radiography of the target segments was divided into two objectives: (1) to obtain a full view or composite shot to verify close spacing of the pellets within the cladding and (2) to verify full penetration of the circumferential welds in the vicinity of the end fittings. The latter was accomplished by taking radiographs of both sides of the weld at 0-, 60-, and 120-deg rotations. This approach provided a total of six views of the weld, which was considered ample to detect gross defects. This reasoning was also predicated on the performance of a continuous and uniform weld, which was a necessary criterion invoked in the quality assurance procedure.

To satisfy the first objective radiographs were taken at a fairly low power setting with brass bars located adjacent to the cladding to reduce scatter. Some of these shots were also acceptable for weld examination; however, in most instances a different technique was needed to obtain a clear delineation of the weld zone and void at the fitting-cladding interface. This technique consisted of placing stainless steel shape corrections, machined to closely fit the contour of the rod, in the weld zone to equalize the beam penetration and reduce scatter. With this arrangement power settings were increased to produce a dark film which was examined with a high-intensity arc light source. Resolution of the weld zone was substantially increased with this procedure; therefore, all subsequent radiographs were taken in an identical manner.

The detection limit, or penetration resolution, was judged to be less than 0.002 in. with the latter technique, and this value was somewhat higher initially. These figures are based for the most part on examinations of penetrometer holes and visibility of the void between the end fitting and cladding wall which extended into the cladding about 0.7 in. All welds were judged to be free of defects and of 100 percent penetration within the limits of detection.

SHIPMENT AND ASSEMBLY

Early in the program a decision was made to ship the target rods to the reactor site in a total of four segments (two for each rod) and to carry out final assembly in the pool side area. A bench consisting of closely aligned plywood standoffs on a 10-ft-long steel channel was constructed to facilitate final joining of the rod sections and loading into a reactor fuel assembly. The entire unit comprising the eight segments and flow mixer was then assembled at BCL prior to shipment.

The three lower segments of each rod, i.e., Segments A, B, and C, were joined according to plan at the laboratory. This operation involved the following steps: (1) use of a torque wrench to tighten adjoining segments, (2) drilling and reaming of a hole through the assembled fittings, (3) pin insertion, and (4) welding of the peripheral surface of the pin to the fitting. A photograph of the assembled target rods fastened to the bench is shown in Figure 18. Note the modified flow mixer assembly at the far end of the target rods.

The two rods were then disassembled into the four remaining segments and transported to Connecticut Yankee Reactor together with the flow mixer, alignment bench, and other tools needed to complete the assembly operation. The rods segments were joined at the reactor pool area without difficulty and loaded into an appropriate fuel assembly for storage until reactor reloading.

The fuel assembly containing the neptunium target rods was loaded into Connecticut Yankee Reactor on May 8, 1971. These targets will undergo continuous irradiation until June, 1972, at which time they will be removed for cooling and subsequent examination at BCL.

DISCUSSION AND CONCLUSIONS

As a result of the studies conducted on this program for the preparation and characterization of $\text{NpO}_2\text{-CaO-ZrO}_2$ target rods for Connecticut Yankee Reactor, information and data have been generated in a variety of areas.

Wet ball milling, used to blend the NpO_2 and calcia-stabilized ZrO_2 powders, was found to be most suitable for mixing these powders of widely different densities.

FIGURE 18. NEPTUNIUM TARGET RODS ASSEMBLED AND FASTENED TO BENCH
Note modified flow mixer (far right) attached to the
top end of the rods.

A sintering study conducted with pellets cold pressed from wet-milled, dry-blended, and as-received powders demonstrated the greater activation of the wet-milled powder and enhanced densification through sintering that could be obtained through particle reduction. A 2-hr sinter in high-purity nitrogen at 1675 C was found to give satisfactory densities for the 15.18, 28.06, and 48.76 w/o NpO_2 compositions; a temperature of 1600 C was used for the 77.22 w/o NpO_2 pellets.

Dry centerless grinding of the sintered pellets was incorporated into the fabrication process to insure uniform clearance between the pellets and the cladding wall.

Chemical analysis studies performed on selected pellets showed that total gas, moisture, and hydrogen contents were considerably less than specified tolerances.

Metallographic examination revealed that the grain size for various batches of pellets was quite uniform, considering the wide range of compositions prepared. Average values determined from the photomicrographs varied from 8 to 10 μ . This factor, in conjunction with the uniformity of observed porosity in all specimens, led to the conclusion that the distribution of the neptunia and calcia-stabilized zirconia was homogeneous. Autoradiographs were taken that generally verified the metallographic observations, providing further evidence of the homogeneous nature of all the pellets.

The linear thermal expansion coefficient of the 15.18 w/o NpO_2 material was found to be 8.2×10^{-6} per C. Thermal cycling tests performed on various pellets confirmed the conformance of the materials to design specifications and their stability to fracture during irradiation.

A document was written detailing complete specifications for the neptunia-zirconia target pellets.

During the course of the program, full quality assurance procedures were developed and conducted for cladding examination, pellet fabrication, pellet characterization, some physical property measurements, rod segment welding, shipping, and final assembly and welding at the reactor site.

ACKNOWLEDGMENTS

The authors would like to express their appreciation for the cooperation received from the many persons involved in various phases of the program. Particular thanks are due to Messrs. Ken Roach and William Mullett of Southern Nuclear Engineering for their guidance and support throughout the effort. The technical assistance provided by Messrs. J. B. Williamson and Robert Hawk of Battelle's Columbus Laboratories contributed extensively to the successful completion of the program according to schedule. Credit is also due Mr. Rudy Grube of the Yankee Atomic Electric Company and Messrs. Bob Traggio and Wes Hartley of the Connecticut Yankee Atomic Power Company. Timely suggestions from Mr. Mike Gaitanis, U.S. AEC, who acted as technical monitor for the program were of considerable aid toward implementation of the overall effort.

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APPENDIX

BATTELLE-COLUMBUS SPECIFICATION FOR
NEPTUNIA-CALCIA-STABILIZED ZIRCONIA TARGET PELLETS

APPENDIX

BATTELLE-COLUMBUS SPECIFICATION FOR
NEPTUNIA-CALCIA-STABILIZED ZIRCONIA TARGET PELLETS

1.0 Scope

- 1.1 This specification applies to solid cylindrical NpO_2 -(CaO - ZrO_2) target pellets that will be encapsulated and irradiated in the Connecticut Yankee Reactor Core III of the Connecticut Yankee Atomic Power Company (CYAPC).

2.0 Design Requirements

- 2.1 No change shall be made in the design details or quality of material furnished under this specification without the written approval of Southern Nuclear Engineering (SNE).

3.0 Manufacture

- 3.1 The pellets shall be prepared from neptunium dioxide (NpO_2) powder supplied by Savannah River and reactor-grade calcia-stabilized (CaO) zirconia (ZrO_2) containing 10 to 12 w/o CaO purchased from ZIRCOA. Standard cold-pressing and sintering techniques will be used for all pellet fabrication. No materials shall be added to the powders except small quantities of organic binders and/or an organic lubricant needed for pelletization.

4.0 Materials Accountability

- 4.1 Accountability and accounting for the NpO_2 shall be in accordance with AEC Manual Chapter 7400. Battelle-Columbus is an Exempt AEC Contractor operating under the contract cognizance of the Chicago Operations Office. Battelle-Columbus will be responsible to the AEC through the Chicago Office for the receipt, processing, removals and losses of the NpO_2 through established accountability and accounting procedures. At the conclusion of the program, a closed accountability balance of all materials received will be furnished to both SNE and the AEC. Any generated scrap or unused materials will be shipped for reprocessing or burial as instructed by the AEC.

5.0 Chemical Characterization and Tests

- 5.1 Neptunium contents of the pellets for the various target segments will conform to those values specified below, which are based on the information given in SNE Drawing No. 69-646-10.

Segment		Neptunium	Total Volume	Total Mass Neptunium, g	Total Mass NpO ₂ , g	NpO ₂ Composition of Target Pellets, w/o
		Density, g per cm ³	of Fuel Segment, cm ³			
1A	2A	5.44	18.88	102.69	116.55	77.22
1B	2B	2.72	18.88	51.35	58.28	48.76
1C	2C	1.36	18.88	25.67	29.13	28.06
1D	2D	0.68	81.17	55.20	62.65	15.18

5.2 Chemical analysis for neptunium, zirconium, and calcium will be performed in accordance with established procedures for these elements.

5.3 Since the oxygen-to-metal ratio of the pellets is unknown and would be meaningless unless referenced to back-up data on phase relationships and stoichiometry, emphasis for pellet characterization and behavior will be placed in microstructural examinations and prolonged heat treatment of encapsulated pellets. Stoichiometry of the pellets will be that which results from sintering to high temperatures in an atmosphere of high-purity nitrogen and is expected to vary slightly for each composition because of the changes in NpO₂ and ZrO₂ contents.

5.4 Impurities present in the pellets shall not exceed the levels given below:

<u>Impurity Element</u>	<u>Maximum Content, ppm</u>
Silver	0.3
Bismuth	2.0
Boron	1.0
Cadmium	5.0
Chromium	200.0
Copper	25.0
Iron	500.0
Lithium	0.5
Magnesium	300.0
Manganese	150.0
Molybdenum	50.0
Nickel	100.0
Lead	60.0
Silicon	300.0
Tin	40.0
Vanadium	20.0
Zinc	50.0

5.5 Spectrographic analysis will be utilized to determine trace impurities.

- 5.6 The hydrogen content of the pellets shall not exceed 10 ppm, including the amount contributed by water. Hydrogen content shall be determined from mass spectrographic analysis of a gas sample taken during measurements for total gas in Paragraph 5.9, below. It shall be assumed that the ratio of hydrogen to other species found in this gas sample is for all practical purposes the same, and this ratio will be used to calculate the hydrogen content of pellets analyzed for total gas release.
- 5.7 Chlorine content shall not exceed 10 ppm as determined from an analysis of a pellet randomly selected from a batch of pellets containing 15.18 w/o NpO_2 .
- 5.8 Total water content shall not exceed 10 ppm. Moisture desorption temperatures employed in the analysis shall be 1000 C.
- 5.9 Total gas release inclusive of water shall not exceed 0.05 cm^3 per g of target material at STP. Gas release shall be measured by outgassing for at least 30 min at 1×10^{-6} mm of mercury at a temperature of 1000 C.
- 5.10 Pellets of each composition will be sectioned and examined metallographically to magnifications of 500 X. This examination shall indicate a minimum of 90 percent solid solution of the NpO_2 -calcia-stabilized ZrO_2 powders.
- 5.11 Pellets examined metallographically will be subjected to quantitative alpha autoradiography. The uniformity of the neptunium dispersion shall be such that there are no areas of high NpO_2 concentration exceeding 200 μ .
- 5.12 One pellet of each composition will be encapsulated in a Type 304 stainless steel can and heat treated for 1 month at a temperature of 500 C. The cladding and pellets will be examined for diffusion interaction of the various constituents prior to the loading of the targets into the reactor.

6.0 Physical Properties and Dimensions

- 6.1 Densities of finished pellets shall be in the range specified below for each target composition.

<u>Segment</u>	<u>NpO_2 Composition, w/o</u>	<u>Total Pellet Density, g per cm^3</u>
1A 2A	77.22	8.60 ± 0.15
1B 2B	48.76	6.10 ± 0.15
1C 2C	28.06	5.40 ± 0.15
1D 2D	15.18	5.10 ± 0.15

- 6.2 Three pellets shall be selected from the pellets prepared from each batch of material and measured for weight and volume. Geometric densities determined from these measurements shall fall within the range specified in Paragraph 6.1, above.

- 6.3 Pellet dimensions shall conform to those given on SNE Drawing No. 69-646-1. Tolerances on the pellets are 0.380 to 0.385 in. for the diameter and 0.350 to 0.650 in. for the length.
- 6.4 The plane of each pellet end shall not deviate from a plane perpendicular to the center axis by more than 0.08 in. across the pellet diameter.
- 6.5 The pellets shall be subjected to visual inspection for chips and fissures and they shall conform to the following specifications.
 - 6.5.1 Chipped pellets shall not have lost more than 10 percent of the area at either end.
 - 6.5.2 The sum of the circumferential length and depth of each imperfection on the cylindrical surface shall not exceed 0.20 in. The maximum axial length of any one chip shall not exceed 0.20 in. The sum of the circumferential lengths of all chips and pock marks in any one plane perpendicular to the pellet axis shall not exceed 0.20 in. In cases of "borderline" acceptability, chipped or fissured pellets may be accepted when they are within the weight per unit volume specification and withstand subsequent handling and tube loading without further deterioration.
- 7.0 Test Sampling
 - 7.1 Dimensional samples
 - 7.1.1 Each pellet shall be inspected visually for chips, fissures, and other imperfections.
 - 7.1.2 All pellets shall be inspected for compliance to dimensional tolerances as stated in Paragraph 6.3.
 - 7.1.3 For pellet squareness, a portion of the pellets may be randomly sampled. The sampling plan and rejection criteria shall be subject to mutual agreement.
 - 7.2 Chemical samples
 - 7.2.1 A batch of material shall be defined as the NpO_2 and CaO -stabilized ZrO_2 powders blended initially by wet ball milling in the amounts necessary to obtain the NpO_2 compositions specified in Paragraph 5.1. A batch of pellets shall consist of the compacts pressed and sintered under identical conditions in the same furnace cycle.
 - 7.2.2 One pellet prepared from each batch of material shall be selected and stored as an archive sample to be utilized during postirradiation evaluation of the target rods.
 - 7.2.3 An analysis is to be performed on one pellet from each batch of material to assure compliance with Section 5.0.

7.2.4 Gas release will be determined using the entire pellet selected at random in Paragraph 7.2.3.

7.2.5 Sections from one pellet from each pellet batch will be examined metallographically and submitted for quantitative autoradiography.

7.2.6 One pellet of each composition will be encapsulated in a Type 304 stainless steel can and heat treated for 1 month at 500 C.

8.0 Test Reports

8.1 Copies of the chemical analyses reported for total neptunium, zirconium, calcium and impurities together with the analyses taken on the cladding and end component material plus reports of physical properties and dimensions shall be submitted to SNE and CYAPC on completion of the program.

9.0 Packaging and Handling

9.1 The pellets shall be packed and handled in a manner that will insure their compliance with the above specification when loaded into the cladding tubes.

9.2 The identity of pellet batches shall be retained throughout the manufacture of finished fuel assemblies.

10.0 Inspection

10.1 The manufacturer shall afford SNE and CYAPC representatives all reasonable facilities, information, and right of inspection to satisfy themselves that the pellets furnished are in accordance with this specification.

10.2 Acceptance of pellets and target rods by SNE or CYAPC shall release Battelle-Columbus of all responsibility for performance of these rods while in the reactor.

11.0 Compliance with Regulations

11.1 The vendor shall have appropriate licenses and clearances required for compliance with all applicable regulations of Federal, State, and Local bodies with respect to receiving, accounting for, processing, storing, and shipping neptunium.

- P5 - PRIMARY SECONDARY NEUTRON SOURCE
 S5 - SECONDARY NEUTRON SOURCE
 () - POSSIBLE CONTROL ROD LOCATION (6 PLACES)
 (X) - ACTUAL CONTROL ROD LOCATION (45 PLACES)
 [] - TARGET ROD ASSEMBLY LOCATION (1 PLACE)

CORE CROSS SECTION
 CONNECTICUT YANKEE REACTOR
 SCALE 3/8"=1'-0"

FUEL ASSEMBLIES J-13, J-14 & J-15
 NOT TO SCALE

NEW FUEL ASSEMBLY J-14 WITH TARGET ROD ASSEMBLY
 NOT TO SCALE

TARGET ROD ASSEMBLY
 SCALE 6"=1'-0"
 SECTION A-A

TARGET ROD #	GM% Np / CM ³ PELLET	VOL Np CM ³	MASS Np GM%
1A 2A	7.44	18.88	102.69
1B 2B	2.72	18.88	71.97
1C 2C	1.96	18.88	29.67
1D 2D	0.68	81.17	77.20

DETAIL 'A'
 (TYP 2 PLCS) SCALE 2:1

NOTE	ITEM NO.	TITLE	SNE DWG & GROUP OR ITEM NO.	MATERIAL SPECIFICATION	NO REQ'D GROUP 1
	1	TARGET ROD ASSEMBLY	69-646-10, GRD		
	2	MODIFIED FLOW MIXER ASSY	69-646-11, IT 1		1
	3	TARGET ROD-UPPER SECTION	69-646-11, IT 1		2
	4	TARGET ROD-LOWER SECTION	69-646-11, IT 2		2
B	5	LOCKING PIN		ASTM A 276, TYPE 304 CENT GRD, COND A	4
	6	FUEL ASSEMBLY (J-14)	WEST DWG ED 36 3/16/67		

NOTE B CARBON CONTENT 0.4% TO 0.6%
 COBALT CONTENT 1% MAX

NOTES:

- CLEAN ALL PARTS BEFORE AND AFTER ASSEMBLY. ALL SURFACES TO BE FREE OF BURRS.
- WELD PER MUTUALLY AGREED UPON PROCEDURE BETWEEN BME & SNE ENGINEERS. VISUALLY INSPECT AT 7X MAGNIFICATION. NO CRACKS PERMITTED.
- TARGET AND SEGMENT IDENTIFICATION NUMBERS MUST MATCH IDENTIFICATION NUMBERS PER TABLE I, THIS DWG.
- THE TARGET ROD ASSEMBLY MUST NOT BE LIFTED FROM THE HORIZONTAL TO THE VERTICAL POSITION WITHOUT BEING SUPPORTED & CLAMPED AT TARGET SUPPORTS OF THE FINAL ASSY & UPENDING FIXTURE. THE DWG 69-646-12, GR 2.
- AN INSPECTION PLUG 7/16 MIN DIA x 1 1/2 LG MUST BE ABLE TO PASS THROUGH THE TWO TARGET ROD THIMBLES OF ITEM #6 A DISTANCE OF 107.00 MIN AND A PLUG 4/81 MIN DIA x 1 1/2 LG MUST BE ABLE TO ENTER 112.00 MIN. PRIOR TO INSTALLATION OF ITEM 1.

RELEASED FOR MANUFACTURE
 12/9/70

REFERENCE DWGS
 SNE: 69-646-11
 SNE: 69-646-12
 REFERENCE SPEC
 SNE SPEC: 69-646-1

C	6-2-71	CHANGED NOTE 5, REDESIGNATED FUEL ASSY TO 100 AND REDESIGNATED DESIGNATION 12
B	12-9-70	CHANGED FUEL LENGTH TO 120.00, MADE CORRESPONDING DIMENSIONAL ADJUSTMENTS
A	11-16-70	MINOR DIMENSIONAL CHANGES
REV	DATE	DESCRIPTION
SNE SOUTHERN NUCLEAR ENGINEERING, INC. DUNEDIN, FLORIDA PROJECT PU 238 COMMERCIAL PRODUCTION TITLE NEPTUNIUM TARGET ROD ASSEMBLY IN FUEL ASSY (J-14) OF CONN. YANKEE REACTOR SCALE 1/4"=1'-0"		
CHECKED	11-2-70	R. COLE
DESIGNED	11-16-70	W. J. C.
APPROVED	12-9-70	M. G. RING
SCALE	1/4"=1'-0"	
NO	69-646-10 C

ITEM NO.	TITLE	SNE DWG & GROUP OR ITEM NO.	MATERIAL SPECIFICATION	NO RE
1	TARGET ROD UPPER SECTION (TAR, ET 'D')	69-646-1, GR 1		
2	TARGET ROD LOWER SECTION (TAR, ET 'D')	69-646-1, GR 2		
3	TARGET ROD - SEGMENT 'A'	69-646-1, GR 3		1
4	TARGET ROD - SEGMENT 'B'	69-646-1, GR 4		1
5	TARGET ROD SEGMENT 'C'	69-646-1, GR 5		1
6	UPPER CONNECTOR	ASTM A 14 TYPE 304 CENT GRD COND A		1
7	TARGET TUBE (LONG)	TYPE 304 77 PER MINUM SPEC UP 84 DIA (SEE NOTE 4) 8 1/2 DIA (SEE NOTE 4) 8 1/2 DIA (SEE NOTE 4) 8 1/2 DIA (SEE NOTE 4)		1
8	TARGET TUBE (SHORT)	TYPE 304 77 PER MINUM SPEC UP 84 DIA (SEE NOTE 4) 8 1/2 DIA (SEE NOTE 4) 8 1/2 DIA (SEE NOTE 4) 8 1/2 DIA (SEE NOTE 4)		1
9	SPRING (LONG)	ASTM A 228 SPRING TIL OIL TEMPERATURE WIRE		1
10	SPRING (SHORT)	ASTM A 228 SPRING TIL OIL TEMPERATURE WIRE		1
11	LOWER CONNECTOR	ASTM A 14 TYPE 304 CENT GRD COND A		1
12	LOWER END FITTING	ASTM A 14 TYPE 304 CENT GRD COND A		1
13	LOCKING PIN	ASTM A 14 TYPE 304 CENT GRD COND A		2
14	ZrO ₂ HALF PELLET	SEE SPEC 69-646-1		2
15	NpO ₂ PELLET (TYPE 'A')	SEE SPEC 69-646-1		2
16	NpO ₂ PELLET (TYPE 'B')	SEE SPEC 69-646-1		2
17	NpO ₂ PELLET (TYPE 'C')	SEE SPEC 69-646-1		2
18	NpO ₂ PELLET (TYPE 'D')	SEE SPEC 69-646-1		2

NOTE A: CARBON CONTENT - 0.4% TO 0.8%
COBALT CONTENT - 0.8% MAX
NOTE B: CARBON CONTENT - 0.4% TO 0.8%
COBALT CONTENT - 1.9% MAX

NOTES:

- ALL SURFACES TO BE FREE OF NICKS AND BURRS.
- PRIOR TO ASSEMBLY, TUBE END CONNECTORS, END FITTINGS & ALL INTERNAL SURFACES & COMPONENTS MUST BE CLEAN & DRY AND FREE OF DUST OR OILY CONTAMINATION. PRIOR TO WELDING, TUBE, END CONNECTORS & FITTINGS MUST BE CLEAN AND DRY.
- WELD PER MUTUALLY AGREED UPON PROCEDURE BETWEEN BNL & SNE ENGRS. NO FILLER WIRE REQUIRED.
- MAXIMUM DIA AFTER WELDING. ALL SURFACES IN THE WELD AREA MUST HAVE A SMOOTH TRANSITION. GRINDING OF TUBE WALL PERMITTED. TOLLING OF WELD PERMITTED PROVIDED THE OPERATION IS RESTRICTED TO THE WELD ZONE AND DOES NOT EXTEND TO THE CLADDING IN THE UNSUPPORTED SECTION. TOLLING PROCEDURE TO BE APPROVED BY SNE.
- EXTERNAL SURFACE OF WELDED AREA MUST BE THOROUGHLY CLEAN. NO RESIDUE OF CHLORINATED COMPOUND ARE ALLOWED. ALL CLEANING PROCEDURE MUST BE SUBMITTED FOR THE APPROVAL.
- TUBE ASSEMBLY MUST BE STRAIGHT WITHIN .010 PER FOOT WHEN LYING ON SURFACE PLATE. END CONNECTORS AND FITTINGS TO BE STRAIGHT WITHIN .010.
- WELD PER MUTUALLY AGREED UPON PROCEDURE BETWEEN BNL & SNE ENGRS. VISUALLY INSPECT WITH 9X MAGNIFICATION. NO CRACKS PERMITTED.
- IF ALL OVER UNLESS OTHERWISE SPECIFIED.
- SPRING MUST PASS FREELY THROUGH AN INSPECTION ENVELOPE OF .787 I.D. MAX X L₀ LONG WITHOUT RESTRICTION OR BINDING.
- INTERPRET GEOMETRIC TOLERANCES PER MIL-STD-88.

DESIGN SPECIFICATIONS	ITEM ⑩	ITEM ⑪
FREE LENGTH (L ₀) (IN)	3.025	3.025
INITIAL DEFLECTION (IN)	714	092
INITIAL LENGTH (IN)	2.700	770
INITIAL LOAD @ INITIAL DEFLECTION (LB)	2.7	1.0
EXPECTED WORKING DEFLECTION (IN)	2.67	062
EXPECTED FINAL DEFLECTION (IN)	841	194
EXPECTED FINAL LOAD (LB)	766	166
MAX SOLID HEIGHT (IN)	1.195	0509
SPRING RATE (LB/IN)	4.35	10.80
TOTAL NUMBER TURNS	27	12
ACTIVE NUMBER TURNS	27	10
STRESS AT INITIAL LOAD CORRECTED (PSI)	76,200	14,600
DIRECTION OF COIL	R H	R H
TYPE OF ENDS	CLOSED & GROUNDED	CLOSED & GROUNDED

REFERENCE DATA	DESCRIPTION
C	2.1 71 AGED SPRING DESIGN DATA, REV PELLET STACK HEIGHT
B	12.5 70 REV LENGTH OF LOWER END FITTING
A	11.16 70 MINOR DIMENSIONAL CHANGES
REV	DATE DESCRIPTION

SOUTHERN NUCLEAR ENGINEERING, INC.	
APPROVED	DUNEDIN, FLORIDA
REVISOR	REVISOR
PROJECT	PROJECT
DATE	DATE
DESCRIPTION	DESCRIPTION
DESIGNED	DESIGNED
CHECKED	CHECKED
APPROVED	APPROVED
REVIEWED	REVIEWED
DATE	DATE
DESCRIPTION	DESCRIPTION
69-646-10	69-646-11C

Fig. 2

Fig. 3

NOTES:

1. PLUG GAGES MAY BE ATTACHED TO BOTH ENDS OF A SINGLE HANDLE (AS SHOWN) OR TO ONE END OF SEPARATE HANDLES OF THE SPECIFIED LENGTH.
2. PLUG GAGE USED PER NOTE 5 OF SNE DWG. 69-646-10.

SOUTHERN NUCLEAR ENGINEERING, INC
DUNEDIN, FLA.

TARGET ROD THIMBLE
PLUG GAGE

DRAWN
BY RCOLE

SCALE
FULL

DRAWING NO.

CHK'D

DATE
2-24-71

69-646-17

REFERENCE DWG:
SNE: 69-646-10

Fig. 4

Fig. 5 Neptunium-237 mass as a function of fuel exposure

Fig. 6 Plutonium-238 mass as a function of fuel exposure

Fig. 7 Plutonium-239 mass as a function of fuel exposure

Fig. 8 Power density in the target pellets as a function of fuel exposure

Fig. 9 Total fissions as a function of fuel exposure

Fig. 10 Fission gas pressure as a function of fuel exposure

Fig. 11 Shutdown temperature transient - Target A

Fig. 12 Shutdown temperature transient — Target B

Fig. 13 Shutdown temperature transient — Target C

Fig. 14 Thimble - fuel tubing flow test apparatus

Fig. 15 Measured flow vs. Δp through Conn. Yankee C.R. thimble with fuel rod inserted in thimble

Fig. 16 Results of PDQ-7 calculations on initial Connecticut Yankee core. (Quarter-core symmetry assumed)

Fig. 17 Flow chart of data preparation programs for PDQ-7-HARMONY

FUEL CHAINS

BURNABLE POISON

NEPTUNIUM TARGETS

Fig. 18 Depletion chains described in HARMONY

Fig. 19 Connecticut Yankee Reactor - First cycle assembly-wise relative power density as a function of average core burnup

18,292	18,321	18,415	18,596	19,259	18,447	15,495	10,122
18,321	18,347	18,426	18,551	18,498	18,035	15,489	8,873
18,415	18,426	18,477	18,496	18,645	16,788	12,906	
18,596	18,551	18,496	18,723	17,410	15,141	9,212	
19,259	18,498	18,645	17,410	14,393	10,505		
18,447	18,035	16,788	15,141	10,505			
15,495	15,489	12,906	9,212	Assembly Burnup, Mwd/mtU			
10,122	8,873						

Fig. 20 Connecticut Yankee Reactor — First cycle assembly-wise burnup at core average burnup of 16,000 Mwd/mtU

Fig. 21 Connecticut Yankee Reactor—First cycle, critical boron concentration as a function of burnup

1.239 1.191	1.187 1.110	1.285 1.249	1.181 1.127	1.223 1.219	1.121 1.121	0.980 1.003	0.749 0.720
1.187 1.110	1.287 1.243	1.287 1.236	1.275 1.257	1.200 1.184	1.122 1.142	0.988 1.003	0.774 0.783
1.285 1.247	1.287 1.235	X ← $F_{\Delta H}^N$ 1.310 1.285	1.238 1.214	1.174 1.185	1.053 1.067	0.905 0.937	0.663 0.654
1.181 1.124	1.275 1.253	1.238 1.212	1.184 1.196	1.028 1.028	0.984 1.032	0.853 0.896	0.551 0.522
1.223 1.214	1.200 1.178	1.174 1.181	1.028 1.026	1.135 1.221	0.832 0.866	0.775 0.801	
1.121 1.115	1.122 1.135	1.053 1.052	0.984 1.027	0.832 0.864	0.895 0.981	0.499 0.471	
0.979 0.999	0.988 0.998	0.905 0.930	0.853 0.890	0.775 0.798	0.499 0.470	SNE FSAR	
0.749 0.731	0.774 0.782	0.663 0.651	0.551 0.519				

Fig. 22 Indian Point Unit No. 2 — Assembly-wise average power distribution beginning of life, unrodded core