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IRRADIATION PERFORMANCE
OF
PIQUA CORE I
FUEL ELEMENTS

By
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RELEASED FOR ANNOUNCEMENT
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CONTENTS

	Page
Abstract	6
I. Introduction	7
II. Fuel Element Design and Fabrication	9
A. Fuel Element Design	9
B. Fuel Element Fabrication	12
III. Irradiation History	13
A. Element P-1071	13
B. Element P-1091	13
IV. Postirradiation Examination	15
A. As-Received Element	15
B. Outer Process Tube	15
C. Fuel Cylinders	19
1. Visual Examination	19
2. Gamma Scanning and Burnup Analyses	21
3. Dimensional Measurements	21
4. Density Measurements	25
5. Metallographic Examination	25
6. Film Deposits Examination	31
D. Inner Process Tube	32
V. Discussion and Evaluation	37
A. Fuel Dimensional Changes	37
B. Nickel Diffusion Barrier	38
C. Film Deposits	39
VI. Conclusions	41
References	42

TABLES

		Page
1.	Density Changes of Fuel From Piqua Fuel Elements P-1071 and P-1091.....	27
2.	Results of Metallographic Examination of Nickel Diffusion Barriers on Piqua Fuel Elements P-1071 and P-1091.....	28

FIGURES

1.	Piqua Core I Fuel Element	10
2.	PNPF Reactor Power Level, as a Function of Time.....	13
3.	Outside Surface of Outer Process Tube From Fuel Element P-1071, Showing Area Where Film Flaked Off Near Weld Seam	16
4.	Fuel Element P-1091, as Received From Reactor	16
5.	Fuel Element P-1091, After Process Tube Slit in Half	
a.	Overall View	18
b.	Inside Surface of Outer Process Tube, Near Top of Element .	18
c.	Inside Surface of Outer Process Tube, Near Axial Centerline of Element (Film-free areas correspond to positions of fins on fuel cylinders)	18
6.	Typical Views of Clad Fuel Cylinders From Fuel Element P-1071, Showing General "Clean" Nature of Heat Transfer Surfaces	
a.	Cylinder No. 7	20
b.	Junction of Cylinders No. 1 and No. 2	20
7.	Typical Views of Clad Fuel Cylinders From Fuel Element P-1091	
a.	4 in. From Top of Cylinder No. 1	22
b.	2 in. From Bottom of Cylinder No. 1	22
c.	2 in. From Bottom of Cylinder No. 2	22
8.	Axial Burnup Profiles for Piqua Core I Fuel Elements	23
9.	Fuel Cylinder Dimensional Changes for Piqua Element P-1071. . .	24
10.	Fuel Cylinder Dimensional Changes for Piqua Element P-1091. . .	26

FIGURES

	Page
11. Nickel Diffusion Barrier Between Aluminum Cladding and Fuel Alloy on Irradiated Fuel Elements From Piqua Core I	
a. Typical View of As-Polished Reaction Zone	30
b. Defect or Bubble in Aluminum Cladding, With Associated Increased Interdiffusion Between Nickel and Fuel Alloy	30
12. Inner Process Tube and Valve Body Assembly From Fuel Element P-1091	
a. Overall View	34
b. Solid Organic in Inner Process Tube	34
c. Drainage Hole in Side of Valve Body	34
d. Disassembled Valve Body Assembly	34
13. Increase in Outer Diameter of Outer Fuel Cylinders From Piqua Core I Fuel Elements P-1071 and P-1091, as a Function of Burnup	36

ABSTRACT

Destructive examinations were made on two fuel elements from the Piqua Nuclear Power Facility (PNPF) Core I, as part of a continuing fuel performance surveillance program, to determine the effects of the organic-cooled reactor environment on the integrity of the fuel elements, and as an aid in determining the achievable Core I fuel lifetime. The general mechanical condition of the fuel elements, the fuel cylinder dimensional changes, coolant decomposition films on heat transfer surfaces, abnormal coolant decomposition products, and the integrity of fuel-cladding diffusion barrier were the principal areas of evaluation. The peak burnups attained were 0.65×10^{20} and 1.19×10^{20} fissions/cc (1400 and 2550 Mwd/MTU) for Fuel Elements P-1071 and P-1091 respectively. In general, the results of the examinations were very encouraging, in that the fuel elements were in excellent mechanical condition. Fuel dimensional changes were small ($<0.8\%$ outer diameter change); surface films on heat transfer surfaces were thin (a maximum thickness of 0.0017 in.); and the fuel-cladding diffusion barrier had maintained its integrity. Decomposed organic coolant deposits were found in the low-flow regions of the inner process tube and in the valve body assembly below it.

I. INTRODUCTION

The Piqua Nuclear Power Facility (PNPF), which was designed and built by Atomics International for the United States Atomic Energy Commission, is a part of the AEC's Power Demonstration Reactor Program. This reactor, which is organic cooled and moderated, is designed to produce 45.5 Mw of thermal energy and 11.4 Mw of electrical power.

Atomics International is conducting a fuel surveillance program to obtain information needed to:

- 1) Maintain safe operation of the reactor
- 2) Determine the useful lifetime of the fuel elements
- 3) Determine the extent of film formation on the heat transfer surfaces
- 4) Determine the limiting mechanical and metallurgical conditions
- 5) Improve further Piqua fuel element designs
- 6) Optimize the fuel management schemes.

The specific data needed for a comprehensive evaluation of the Piqua Core I fuel elements will be compiled through a program which incorporates the following major tasks:

- 1) Detailed preirradiation dimensional measurements of six standard and four instrumented fuel elements
- 2) Hot cell examination of irradiated fuel elements, including dimensional measurements, metallography, gamma scans, and burnup analysis
- 3) Analysis of irradiated fuel element surface conditions, as related to heat transfer characteristics
- 4) Analysis of experimental data obtained, with the objectives of providing an optimum fuel management scheme and of making recommendations for the design of improved fuel element assemblies.

To date, two fuel elements from Piqua Core I have been removed from the reactor and examined at the Atomics International Hot Laboratory (AIHL). This report covers the postirradiation examination and evaluation of these

fuel elements, and relates their performance to that expected of the remaining elements in the core. The first element examined (P-1071) was a special, pre-measured fuel element which had a peak burnup of 0.65×10^{20} fissions/cc (1400 Mwd/MTU). The second evaluation element was scheduled to be removed after a peak burnup of $\sim 1.19 \times 10^{20}$ fissions/cc (2550 Mwd/MTU). However, as the time approached for the second examination, another element (P-1091) began to exhibit an unexplained steady increase in coolant exit temperature ($\sim 40\%$ total increase). Although this element was not one of the ten premeasured elements, it was selected for removal and examination, because of interest in this temperature rise. This did not significantly affect the overall surveillance program, since much of the information sought on evaluation elements could be obtained as well from nonmeasured elements as from premeasured ones, namely:

- 1) The extent and nature of surface film formation
- 2) The extent of fuel-cladding nickel bond consumption
- 3) Dimensional and density changes of the fuel cylinders
- 4) General mechanical stability of the fuel assembly.

Of these, only Item 3 was affected by substitution of a standard element for a premeasured element, and even this item could be determined fairly well by statistical means.

II. FUEL ELEMENT DESIGN AND FABRICATION

A. FUEL ELEMENT DESIGN

The fuel elements consist of a metallic uranium alloy, clad with aluminum. The alloy is composed of uranium enriched to 1.94 wt % in U^{235} , with 3.5 wt % molybdenum and 0.1 wt % aluminum as stabilizing elements. The aluminum cladding, which has a finned surface to increase the heat transfer area, is metallurgically bonded to the uranium fuel, using an intermediate nickel diffusion barrier with a thickness of 0.0010 to 0.0015 in. A cutaway view of the fuel element is shown in Figure 1.

The fuel elements are circular in cross section, and have an overall length of 80.5 in. One fuel element contains four outer fuel cylinders and four inner fuel cylinders, stacked in two concentric tiers. Each fuel cylinder is 13.6 in. long, with the outer cylinders having a mean diameter of 4.63 in. and the inner cylinders having a mean diameter of 3.60 in. The fuel alloy in each cylinder is 0.208 in. thick, and is covered on the inner and outer surfaces with finned aluminum cladding. The aluminum cladding in each end of the fuel cylinder is machined to provide proper nesting of the cylinders when stacked.

The fins in the surface of the Type 1100 aluminum cladding have a spiral of 30°/ft; the rotational direction of the spiral on the inside surface of the outer cylinder is opposite that on the outside surface of the inner cylinder, in order to increase the reactor heat transfer performance and to promote coolant mixing. Initial contact of the fin tips is expected, and has no consequence, because of the high thermal conductivity of the aluminum and the spiralling nature of the coolant flow.

The inside of the inner cylinders and the outside of the outer cylinders are surrounded by 0.010-in. thick AISI Type 347 and Type 304 stainless steel process tubes, which are used to confine the coolant flow and increase its velocity through the elements. The inner process tube has a 3.08 in. ID, and the outer process tube has a 5.21 in. OD. The ends of the outer process tube are welded to the upper head assembly and the lower plug assembly, so that the weight of the fuel cylinders is transmitted by the outer process tube to the

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Figure 1. Piqua Core I Fuel Element

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upper head assembly, which rests on the upper grid plate. The fuel elements are always handled by an attachment to the upper head assembly, so that the outer process tube is loaded in tension.

Two stainless steel screens are welded to the inner process tube, one located above the fuel cylinders at the entrance to the annulus between the process tubes, and the other located above the "weep hole" at the bottom of the inner process tube. The upper screen, which prevents the entry of large particles into the coolant channels of the fuel cylinders, also reduces the possibility of hot spots. The screen at the bottom of the inner process tube prevents the entry of particles into the weep hole which allows only a limited flow of organic coolant through the inner process tube. Thus, the bulk of the coolant flow is through the flow channels adjacent to the fuel cylinders.

Those fuel elements which are not used as control rod elements contain a variable orifice assembly in the entrance section (upper head assembly). The variable orifice assembly consists of a fixed orifice plate, a tapered plug mounted concentrically with respect to the orifice on a threaded shaft, and a locking washer. The flow through the element is varied remotely by a screw mechanism which produces a nearly linear flow variation as a function of the number of turns. The approximate range of flow adjustment that can be achieved is from 75 to 200 gpm. Complete flow blockage is impossible, since the maximum diameter of the plug is smaller than the orifice plate opening. A locking device automatically secures the screw adjustment mechanism as the adjusting tool is removed.

The function of the inner process tube is to direct organic coolant through the flow channels adjacent to the fuel cylinders. There is a small orifice hole (7/64 in. diameter) in the side of the valve body at the bottom of the inner process tube. This hole allows a small flow of organic through the inside of the inner process tube during reactor operation, and provides a means of removing the heat generated in this central moderator region. Except for this hole, the valve body is essentially closed when in position in the reactor core. However, when the fuel element is lifted out of this position, such as during removal of an element from the core, a ball plug on the center shaft is designed to fall out of its position inside the valve body. This allows the coolant to drain

from the inner process tube at a faster rate, and results in a minimum delay in removal of the element from the core.

B. FUEL ELEMENT FABRICATION

The fuel alloy was prepared by vacuum melting the enriched uranium metal with the alloying metals (3.5 wt % molybdenum and 0.1 wt % aluminum) and casting it into cylinders in graphite molds. The cast fuel cylinders were then cut to length and machined to final size. After the machining operation, the cylinders were ultrasonically cleaned and vacuum outgassed at 1000°F. The cleaned, outgassed fuel cylinders were nickel plated, and were tested for proper thickness of nickel by an eddy-current testing device. Nickel film thicknesses were maintained between 0.0010 and 0.0015 in.

The cleaned, nickel-plated fuel cylinders were then placed in clean aluminum cladding extrusions, and aluminum end plugs were welded into place on each end. Bonding of the aluminum cladding to the fuel cylinders was accomplished by heating the assembly to 1000°F and applying an argon gas pressure of 7000 psi. After final machining of the clad fuel cylinder, they were placed in a steam autoclave and tested at 250°F and 15 psi. Any crack in the cladding would allow the steam to penetrate the cladding, and would cause a blister at that point. This provided a means of testing the integrity of the aluminum cladding. To test for the metallurgical bond integrity between the fuel alloy and the aluminum cladding, a thermal transient technique was used. The technique consisted of cooling the internal surfaces of the cylinder with water while the outer surface was heated with a gas flame and scanned with an infrared detector. During this process, the fuel cylinder was rotated, and the infrared detector was advanced 1/4 in. per revolution. The infrared detector was focused on the fuel element's outer surface, 90° from the point of impingement of the flame. Any unbonded area between the nickel and the aluminum or uranium, either on the inner or outer surfaces of the fuel cylinder, would appear to the detector as a hot spot, since the unbonded areas act as thermal barriers and decrease the ability of the cooling water to remove the heat.

During the first period, the relative power of this element remained essentially constant at 1.35. At this time, a special dummy element was removed from the position and replaced with a standard driver element. The dummy element was replaced with a standard driver element. The first evaluation fuel element was removed from Core Position D-9 on June 5, 1964, approximately one year after its insertion (June 7, 1963). This was one of the fuel elements which had received extensive preirradiation characterization. The average and peak burnup values were calculated to be 0.43×10^{20} and 0.65×10^{20} fissions/cc (920 and 1400 Mwd/MTU) respectively. At full reactor power (45.5 Mwt), the peak heat flux would be 105,000 Btu/hr-ft², with a maximum cladding temperature of 720°F. A summary of the reactor operating history is given in Figure 2. The relative power of this fuel element, as determined by coolant channel outlet temperature, remained essentially constant over its entire irradiation life. (Relative power is defined as the ratio of the power output of a given fuel element to the average power output of all the fuel elements.)

Figure 2. PNPf Reactor Power Level, as a Function of Time

B. ELEMENT P-1091

The second fuel element examined from Piqua Core I was removed from Position F-13 on December 12, 1965, 15 months after its insertion (September 5, 1963). This fuel element did not receive a detailed preirradiation characterization, but all its dimensions were within the manufacturing specification

limits. During the first period of operation, to May 20, 1964, the relative power of this element remained essentially constant, at 1.35. At this time, a special dummy element was removed from the position adjacent to the P-1091 element. The dummy element was replaced with a standard driver element. As a consequence of this rearrangement, the relative power of P-1091 was increased to 1.54. This relative power remained nearly constant for slightly less than one month; it then apparently increased steadily, to a final relative power of 2.25, just before its removal. This behavior was believed to reflect a progressive film buildup and flow restriction within the coolant channel, with a corresponding increase in coolant channel outlet temperature. This condition necessitated a reactor power level restriction (~ 32 Mwt), to assure that the technical specification limits on maximum heat flux were not exceeded. Although the original plan for the fuel element examinations was based on the periodic removal of premeasured elements, this element was substituted to ascertain the cause of the steady increase in the calculated relative power. Based on a relative power of 1.35 for the period to May 20, 1964 and 1.54 for the remaining operating period, the average and peak burnup values were calculated to be 0.79×10^{20} and 1.19×10^{20} fissions/cc (1700 and 2550 Mwd/MTU) respectively.

IV. POSTIRRADIATION EXAMINATION

A. AS-RECEIVED ELEMENT

The postirradiation examinations of the two fuel elements were conducted separately at the Atomics International Hot Laboratory (AIHL). Fuel Element P-1071 appeared to be in excellent mechanical condition, when removed from the shipping cask, and there were no signs of handling or operational damage. Fuel Element P-1091 also appeared to be in good mechanical condition, with the exception of two small, 3/4-in. long dents in the outer process tube. Subsequent examination of the inside surface of the outer process tube revealed that there were no unusual buildups of decomposed coolant associated with these dents, as would be expected if they were present during reactor operation. It was therefore concluded that these dents were the result of a handling operation during or after removal from the reactor, and their presence had no detrimental effect on the operation of the element in the reactor. The upper and lower head assemblies of both fuel elements were also clean and in good condition.

B. OUTER PROCESS TUBE

Outside diameter measurements were taken of the outer process tube on each of the fuel elements, using a remote-controlled commercial pi-tape (calibrated steel tape which encircles the circumference of a cylindrical object and gives diameter measurements directly). The results of these measurements showed that there was essentially no change in the outside diameter of the outer process tube on either of the fuel elements.

A thin black film covered most of the outer surface of the outer process tube on the P-1071 fuel element. This film was fairly uniform over most of the outer surface, except for some flaking which occurred adjacent to the longitudinal weld seam. Figure 3 shows this flaking, which occurred slightly above the axial midplane of the P-1071 fuel element. Metallographically, this film was found to be ~0.0005 in. thick and fairly uniform over most of the surface, being the thickest (~0.0015 in.) near the longitudinal weld. This thicker film flaked from the outer process tube when the tube was flexed, leaving a bright shiny surface exposed. The inside surface of the outer process tube had a very thin, adherent, dark film over all of its surface. The thickness

974C

Figure 3. Outside Surface of Outer Process Tube From Fuel Element P-1071, Showing Area Where Film Flaked Off Near Weld Seam

PQ-2-2

Figure 4. Fuel Element P-1091, as Received From Reactor

of this film could not be measured metallographically, probably because it was so thin and brittle that it did not remain on the inside surface of the outer process tube when samples were mounted for metallographic examination.

The outer surface of the outer process tube on Fuel Element P-1091 had a black, loosely adhering film near the top, with most of the rest of the surface appearing shiny. Apparently, at some time during reactor operation, the film on the outer surface of the outer process tube was thicker in the higher-temperature, high-neutron-flux regions (i. e., nearer the axial centerline and lower portions of the fuel element). However, at the time of the examination, the film deposits in this region had flaked off, probably when the element was removed from the reactor, since most of the surface was very clean and shiny. Figure 4 shows the outer surface of the outer process tube of Fuel Element P-1091, as it was received at the AIHL. A relatively continuous thin black film was observed on the inner surface of the outer process tube, being thicker in the lower half of the element. Figure 5a shows a view of the P-1091 fuel element, immediately after the outer process tube was removed. Closer examination of the upper portion of the inner surface of the outer process tube showed that the film was relatively thin and adherent (Figure 5b), while, in the lower portion, it was considerably more flaky and rough (Figure 5c). Some areas on the inside surface of the process tube had an appearance that suggested that the fins of the fuel cylinders had either contacted the outer process tube, or at least prevented the film deposition in highly localized areas. Figure 5c shows how the heavier film was blistering, to give a "paint remover" appearance. Since these were hollow blisters, it is believed that they did not form in pile, but probably resulted from soaking in the organic liquid (HB-40) after the fuel element was removed from the reactor. If this blistering had occurred in pile, it would have provided null flow regions for the circulating organic coolant, and these regions would have been highly susceptible to both thermal and radiolytic decomposition. Therefore, one would have expected to find solid blisters, full of decomposed organic or coke, instead of the hollow blisters. Metallographically, the thickness of this film was determined to be in the range of 0.0003 to 0.0011 in. with an average thickness of ~ 0.0006 in.

PQ-2-9

a. Overall View

PQ-2-108

b. Inside Surface of Outer Process Tube, Near Top of Element

PQ-2-109

c. Inside Surface of Outer Process Tube, Near Axial Centerline of Element (Film-free areas correspond to positions of fins on fuel cylinders)

Figure 5. Fuel Element P-1091, After Process Tube Slit in Half

C. FUEL CYLINDERS

1. Visual Examination

For the purpose of convenience the outer ring of fuel cylinders will be designated as Cylinders No. 1 through No. 4, top to bottom; while the inner ring of fuel cylinders will be designated as Cylinders No. 5 through No. 8, top to bottom.

The fuel cylinders from Fuel Element P-1071 appeared to be in generally excellent condition. The outer fuel cylinders had a thin, black, tightly adhering film over most of their surfaces, the film being nearly nonexistent at the top of the fuel column and thickest slightly below the axial centerline of the element. The inner fuel cylinders appeared to have an even thinner film over their outer surfaces. This film was very thin, or nonexistent, for approximately the top 8 in. of Cylinder No. 5, and again was the thickest on Cylinder No. 7. Figure 6 shows the general condition of the fuel cylinders from the P-1071 fuel element.

Visual examination of the fuel cylinders from Fuel Element P-1091 showed that there was a very thin, continuous film on all of Cylinders No. 1, No. 5, and No. 6, and on most of Cylinders No. 2 and No. 4. This film was similar in appearance to that observed on the P-1071 fuel element. The remaining cylinders also appeared to have a thin film; but it had blistered or peeled from the aluminum fins to form hollow blisters, similar to those observed on the inside of the outer process tube. On part of Cylinder No. 8, the film had flaked off of the aluminum fins, leaving behind a lighter colored thin film. None of the coolant channels were blocked on any of the cylinders, and no carbonaceous growths were observed. The film generally was very brittle, and was fairly easily removed with a nylon brush. Again, had this blistering occurred in pile, one would not have expected to find these blisters remaining hollow, for the reasons cited previously. A closer examination of the adherent and the blistered films showed that both had an extremely rough appearance. This large increase in surface roughness could possibly account for a significant increase in the temperature rise through the coolant channel, because of its influence on coolant flow rate (with a constant available pressure drop). However, at the time of these examinations, no means were available to obtain

a. Cylinder No. 7

153D

b. Junction of Cylinders
No. 1 and No. 2

111D

Figure 6. Typical Views of Clad Fuel Cylinders From Fuel Element P-1071, Showing General "Clean" Nature of Heat Transfer Surfaces

quantitative values for the surface roughness of the elements. Typical views, illustrating the general condition of the fuel cylinders from Fuel Element P-1091, are presented in Figure 7.

2. Gamma Scanning and Burnup Analyses

An axial burnup profile was obtained for both of the fuel elements by measuring the gamma activity along the length of the individual fuel cylinders at four angular orientations. This was accomplished by traversing the fuel cylinders in front of a 1/8 by 1/8 by 42 in. orifice through the cell wall, and measuring the Zr^{95} gamma activity coming through this orifice. A relative burnup profile can thus be obtained. In order to obtain the absolute burnup profile, the actual burnup from at least one location must be known. Samples of the fuel were core drilled from the cylinders with an Elox* machine and submitted for radiochemical burnup analyses.

Figure 8 shows the axial burnup profiles for both of the fuel elements. The curves were obtained by averaging the gamma scans taken at the four orientations and correlating them to the peak and average burnups determined by thermal analysis. As can be seen from these curves, there is very good correlation between the data obtained by radiochemical analyses and by thermal analyses for the P-1071 fuel element. The correlation of the value for burnup in the P-1091 element determined by heat transfer with that determined by radiochemical analysis was not as precise as that obtained with the P-1071 element. Therefore, because of the need to provide a reliable prediction of permissible fuel element life, the burnup value obtained from heat transfer data should be used for this prediction. This approach is recommended, because estimates of the burnup achieved can be constantly updated from coolant temperature and flowrate data.

3. Dimensional Measurements

Outer diameter, inner diameter, and length measurements were taken on all of the fuel cylinders, using a pi-tape, tri-mike,† and dial indicator respectively. The aluminum cladding and the nickel diffusion barrier were removed from two

*Elox – electrical discharge method of machining electrically conductive materials.

†Tri-mike – a three-pronged micrometer for taking inner measurements.

a. 4 in. From Top of
Cylinder No. 1

PQ-2-76

b. 2 in. From Bottom of
Cylinder No. 1

PQ-2-74

c. 2 in. From Bottom of
Cylinder No. 2

PQ-2-86

Figure 7. Typical Views of Clad Fuel Cylinders From Fuel Element P-1091

4-22-66

7615-5103

Figure 8. Axial Burnup Profiles for Piqua Core I Fuel Elements

Figure 9. Fuel Cylinder Dimensional Changes for Piqua Element P-1071

fuel cylinders of each element, and the bare fuel dimensions were also obtained. All of the fuel cylinders from Element P-1071 had been measured prior to their insertion into the reactor, both before and after the aluminum cladding had been applied. Therefore, the determination of the dimensional changes on this element consisted simply of comparing the pre- and post-irradiation measurements taken at specific locations on each of the individual cylinders. A different approach was required with Element P-1091, since it was not one of the ten fuel elements which had been selected for extensive preirradiation characterization. However, it had met the manufacturing specifications for the elements fabricated for Piqua Core I. An assumption was therefore made that the 80 fuel cylinders for the 10 premeasured fuel elements were representative of all of the fuel cylinders fabricated, and that Fuel Element P-1091 had fuel cylinders which also were typical of all of the fuel cylinders fabricated. This assumption was supported by a check of the manufacturing records, which confirmed that the carefully measured fuel cylinders had been selected at random. Using this assumption, a statistical analysis of the data was performed, in order to determine the magnitude of fuel swelling. The fuel cylinder dimensional changes for both the clad and bare fuel are illustrated in Figures 9 and 10 for Fuel Elements P-1071 and P-1091 respectively. The points plotted for the P-1091 fuel element were values obtained by comparing the postirradiation dimensions, while the ranges given represent 95% confidence limits.

4. Density Measurements

Density measurements were performed on two fuel specimens that were core drilled from each fuel element. The Archimedes principle of liquid displacement was used in the measurements of the fuel samples after the aluminum cladding and the nickel diffusion barrier had been chemically dissolved. The results of the density determinations are summarized in Table 1. The preirradiation density of the fuel from Fuel Element P-1091 is an average of the preirradiation densities of all the fuel cylinders which were measured. The range of preirradiation densities was 17.94 to 18.08 g/cc.

5. Metallographic Examination

Six fuel specimens were core drilled from each fuel element for metallographic examination. The basic purpose for examining the fuel specimens was

Figure 10. Fuel Cylinder Dimensional Changes for Piqua Element P-1091

TABLE 1
DENSITY CHANGES OF FUEL FROM PIQUA FUEL ELEMENTS P-1071 AND P-1091

Specimen No.	Distance from Top of Fuel Column (in.)	Burnup		Pre-irradiation Density (g/cc)	Post-irradiation Density (g/cc)	$\Delta\rho$ (%)
		(fissions/cc)	(Mwd/MTU)			
P-1071-A	33 (Cylinder 3)	0.60×10^{20}	1300	17.99	17.93	-0.34
P-1071-B	39 (Cylinder 5)	0.58×10^{20}	1250	17.99	17.91	-0.45
P-1091-A	26 (Cylinder 2)	1.14×10^{20}	2450	18.08	17.94	-0.77
P-1091-B	39 (Cylinder 3)	1.16×10^{20}	2500	18.08	17.98	-0.55

TABLE 2

RESULTS OF METALLOGRAPHIC EXAMINATION OF NICKEL DIFFUSION
BARRIERS ON PIQUA FUEL ELEMENTS P-1071 AND P-1091

Distance From Top of Fuel Column (in.)	Cylinder	Surface	Ni (mils)	Al ₃ Ni ₂ (mils)	Al ₃ Ni (mils)	Ni Consumed (mils)
<u>P-1071</u>						
11	1	outer	1.1	0.5	0.4	0.23
		inner	1.2	0.6	0.3	0.25
11	1	outer	1.3	---	0.4	0.08
		inner	1.3		0.5	0.09
26	2	outer	1.2	0.3	0.6	0.21
		inner	1.3	0.2	0.5	0.16
26	2	outer	1.1	0.3	0.6	0.21
		inner	1.1	0.3	0.5	0.19
11	5	outer	1.03	---	0.4	0.08
		inner	1.3	---	0.4	0.08
32	7	outer	1.1	0.3	0.4	0.17
		inner	2.0	0.5	0.4	0.23
<u>P-1091</u>						
16	2	outer	0.98	0.29	0.41	0.17
		inner	1.33	0.20	0.54	0.17
25	2	outer	0.92	0.41	0.36	0.20
		inner	1.03	0.40	0.36	0.20
40	3	outer	1.05	0.91	0.22	0.33
		inner	0.88	0.91	0.35	0.36
40	3	outer	1.05	0.86	0.43	0.36
		inner	1.10	0.86	0.47	0.36
48	4	outer	1.34	0.48	0.35	0.22
		inner	1.19	0.80	0.32	0.31
40	7	outer	0.98	0.46	0.34	0.21
		inner	1.14	0.37	0.44	0.20

to investigate the general integrity of the fuel-diffusion barrier-cladding system, and to determine whether irradiation had any gross detrimental effect on the diffusion rates expected in this system. Other aspects of this examination included an investigation of the stability of the fuel alloy and the possibility of a fuel-cladding separation during reactor operation. All the specimens were mounted, ground, and polished by standard techniques, with emphasis placed on keeping the various surfaces flat for purposes of phase width measurements.

The thickness of the unreacted portion of the nickel diffusion barrier and the thicknesses of the individual phases formed by the aluminum-nickel reaction were measured for each specimen. The fuel alloy-nickel reaction zones were very thin, and were not measurable on nearly all the specimens examined. Table 2 summarizes the data obtained on the examination of the nickel diffusion barrier on the fuel specimens from the Piqua Core I fuel elements. The amount of nickel that is normally consumed in the fabrication of the fuel elements varies from ~ 0.00008 to 0.00011 in. Since the minimum pre-irradiation thickness of the nickel layer is 0.0010 in., this examination showed that nickel performed successfully as an effective diffusion barrier between the fuel alloy and the aluminum cladding.

The postirradiation microstructure of the fuel alloy was sound and nonporous.

The possibility of the aluminum cladding separating from the fuel during reactor operations was investigated by examining the fuel alloy-nickel interdiffusion layer. If there had been a separation in place, a hot spot would have been produced, resulting in a substantially greater fuel alloy-nickel interdiffusion layer. No excessive interdiffusion of these species was noted on any specimens examined. A few localized regions were found metallographically, where there apparently had been a defect or bubble formed between the aluminum and the nickel during fabrication. These defects were generally <5 mils high and <10 mils long. In these very localized regions, a fuel alloy-nickel reaction zone, ~ 0.2 mils thick, was observed; while, over most of the remainder of the specimen, this reaction product was nearly nonexistent. This demonstrates that, if there had been a general lack of bonding of the aluminum cladding, this reaction zone would have been apparent. A typical view of the nickel diffusion layer is shown in Figure 11a, and a view of the fuel alloy-nickel diffusion zone is shown in Figure 11b.

a. Typical View of As - Polished Reaction Zone

15-4-9

b. Defect or Bubble in Aluminum Cladding, With Associated Increased Interdiffusion Between Nickel and Fuel Alloy

7615-1506

Figure 11. Nickel Diffusion Barrier Between Aluminum Cladding and Fuel Alloy on Irradiated Fuel Elements From Piqua Core I

6. Film Deposits Examination

The film deposits on the fuel cylinders of Fuel Element P-1071 were very thin and tightly adherent. They were almost nonexistent nearer the top and bottom of the element (i. e., in the lower temperature, lower neutron flux regions of the fuel column). Various attempts were made to obtain a value for the thickness of this film on the heat transfer surfaces of the fuel cylinders. A method of determining an average film thickness (by scraping clean a given area with a nylon brush) proved unsuccessful, because the film was so light that it became airborne immediately when it was scraped off. Examining the film on the fuel metallographic specimens showed that the film was discontinuous, indicating that it had been damaged somewhat in handling. The thickness of the film, when detectable, ranged from 0.00006 to 0.00014 in.

Some samples of the film deposits on fuel cylinders of the P-1071 fuel element were obtained for chemical analyses by using a more abrasive wire brush, which actually removed some of the aluminum with the film. Results of these analyses showed that the inorganic components of the film were concentrated, by a factor of $\sim 10^4$ over their concentration in the coolant, and that the film contained 12 to 16 wt % iron. No further analysis of this film was made, due to the presence of the aluminum in the samples.

The film deposits on the fuel cylinders of Fuel Element P-1091 were generally thicker than those observed on the P-1071 element, and it was therefore easier to obtain samples of these films for more detailed examination outside the hot cell. Examination of the films on the fuel metallographic specimens showed again that the film was not continuous, and had a thickness ranging from 0.00015 to 0.00054 in. Data obtained from the films that were removed from the hot cell showed that the thickness of the film generally followed the flux profile of the fuel element, ranging from a low of 0.0004 in. to a high of 0.0010 in. Individual values for film thickness, however, were as great as 0.0017 in. The density of the films was found to increase from the inlet (top) to the outlet (bottom) end of the fuel element, from 1.4 to 1.5 g/cc; while the ash content remained fairly constant, at ~ 5 to 10 wt %. The carbon-to-hydrogen atomic ratio for the films on these fuel cylinders was ~ 2 .

Examination of the film which had blistered into the coolant channels revealed that the blisters were indeed hollow, and not clinker-like deposits. If this blistering had occurred in pile, it would have provided a place for a null flow volume of organic coolant which would have become subject to extensive pyrolytic and radiolytic decomposition. Instead, this film had an appearance similar to that of paint after exposure to paint remover; this suggests that the blistering probably occurred while the element was stored in a static liquid, such as in the reactor storage channel, or in the fuel element shipping cask.

D. INNER PROCESS TUBE

The inner process tubes from both fuel elements showed no change in their outer diameters, as a result of their irradiation exposure. A visual examination showed that both inner process tubes were in good condition, with only a relatively thin continuous film observed on the outer surfaces. However, on both the elements, the valve plungers, which should have been fully extended below the bottom of the valve body, were stuck in the closed position.

The inner process tube from Fuel Element P-1071 was nearly full of a liquid, which was analyzed to be predominantly HB-40 (the liquid organic shipping medium). The inner process tube was cut ~1 in. from the bottom, and a massive piece of solid waxy material, 3 to 4 in. thick, was removed. The remainder of the material that was inside the inner process tube was apparently the organic shipping liquid, HB-40. The solid material that was removed appeared to be a mixture of solidified organic coolant (lighter colored, waxy appearance) and radiolytic and/or pyrolytic decomposition products of the organic coolant (black, particulate material). The concentration of the decomposed organic increased toward the bottom of the inner process tube; and, when the valve body was sectioned, it was found to be almost entirely full of decomposed organic. Apparently, it was the presence of this material that had caused the plunger to stick in the closed position, thereby reducing the flowrate of the organic from the inner process tube when the fuel element was removed from the reactor core. This reduced drainage rate probably was not sufficient to allow complete drainage of the inner process tube, so that the remaining coolant solidified when the fuel element was put into the cooling channel (which contained HB-40). This hypothesis explains the fact that the inner process tube

was full of the liquid HB-40. Chemical analyses of this solid material showed that ash content was ~ 4 wt % in the inner process tube, and ~ 8 wt % in the valve body. Samples of the material from the inner process tube were dissolved in tetrahydrofuran, and insolubles were found to increase from 4 wt % near the top to 22 wt % near the bottom. Both the material in the inner process tube and in the valve body were found to be amorphous by x-ray diffraction analysis.

The inner process tube from Fuel Element P-1091 was nearly full of solidified organic. Near the top, the HB-40 used in the shipping canister had softened the coolant; but, over most of its length, the tube was filled with solid coolant. Upon removing this material from the inner process tube, it was noticed that small black particles were dispersed in the coolant throughout the entire length of the tube. This black coke represented ~ 5 wt % of all the material in the tube. These particles were generally of the size 1 by 1 by 0.5 cm. Some chunks of this material were smooth on one side, indicating that they had formed on the wall of the inner process tube; while others were equally rough on all sides, indicating no wall contact. The ash content of this coke was 0.14 wt %, which is a factor of ~ 50 less than the ash content in the film deposits on the heat transfer surfaces. With the exception of the ash content and the carbon-to-hydrogen ratio, which was found to be slightly > 3 , all of the other properties of the coke were similar to those of the film deposits (i. e., they were nonmagnetic, brittle, amorphous, and of nearly equivalent chemical composition).

A sample of the coolant was taken from a section near the top of the inner process tube and was analyzed for high boilers (HB), to see if after-glow irradiation of the coolant caused any significant increase in the HB content. The HB content was found to be 6.5%, which agreed (within the accuracy of the analysis) with the HB content of the PNPf coolant (6.8%) when the element was removed from the reactor. This indicated no detectable change in the decomposition products in the coolant by postoperation irradiation. This also indicated that the coolant in the inner process tube was constantly being replaced by fresh coolant while the reactor was operating.

The plunger in the valve body at the bottom of the inner process tube of the P-1091 fuel element was found to be stuck in the closed position. The drain hole in the side of the valve body was only partially open, and resulted in a

PQ-2-25

a. Overall View

PQ-2-114

b. Solid Organic in Inner Process Tube

PQ-2-31

c. Drainage Hole in Side of Valve Body

d. Disassembled Valve Body Assembly

PQ-2-122

Figure 12. Inner Process Tube and Valve Body Assembly From Fuel Element P-1091

NAA-SR-11712

very slow drainage of organic. This can be seen as the stained area below the hole in Figure 12c. The valve body was cut open; and a very hard, porous, carbonized material was found to fill most of the space above the ball on the valve stem, as illustrated in Figure 12d. Of particular importance is the fact that some solid organic coolant was also found in the sectioned valve body. This supported the belief that there still was some coolant flow through the inner process tube, even though it was probably reduced substantially below original design conditions. The composition of the solid coked material was similar to that of the solid particles found in the inner process tube.

While restriction of flow through the inner process tube does not have a pronounced effect on the overall heat transfer of a fuel element, there are some obvious problems which could develop:

- 1) The coke could eventually build up in the control rod elements, to a point where the control rods could not be fully inserted.
- 2) Complete flow stoppage could cause higher temperatures inside of the inner process tube, which would decrease the coolant density in this region. Since the coolant is also the primary moderator, its decreased density would decrease the total moderation in the core, which would decrease the total reactivity of the core.
- 3) The inner process tube might act as a trap to collect the glass fibers, which contain 2 to 7% boron, from the primary coolant filtering system. However, chemical and microscopic examination of the samples from the inner process tube did not reveal the presence of these glass fibers. Therefore, neutron poisoning by this method was not a problem in either of these two fuel elements.
- 4) Continuous buildup of the coke could eventually cause excess swelling and internal pressures on the inner process tube, which could cause it to deform or fail.

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7615-5107

Figure 13. Increase in Outer Diameter of Outer Fuel Cylinders
From Piqua Core I Fuel Elements P-1071 and P-1091,
as a Function of Burnup

V. DISCUSSION AND EVALUATION

A. FUEL DIMENSIONAL CHANGES

The determination of the dimensional changes in the fuel cylinders was fairly difficult, because of a combination of the relatively large dimensions and the small dimensional changes. However, the magnitude of the changes that can be tolerated in the present design of the Piqua reactor are also relatively small. At full power operating temperatures, the nominal dimensions allow for 1.2% increase in the outer diameter of the outer fuel cylinder before the cladding fins establish firm contact with the outer process tube. Further outer diameter fuel growth can only be accommodated by creep collapse of the cladding fins or by process tube strain. The fin deformation is considered to be the more likely mechanism. A clearance, equivalent to another 1.8% increase in the outer diameter of the outer fuel cylinder, is available between the outer process tube and the hole in the upper grid plate, through which a fuel element must pass to be removed from the core. This represents a limiting condition, since it would significantly affect the normal fuel removal operations. Excessive flow reduction, caused by a decrease in flow area, would represent another limitation which might be imposed by the fuel cylinder dimensional changes. This flow reduction would cause proportionately higher temperatures in the fuel element, which could have several harmful effects:

- 1) Increased fuel-barrier-cladding interdiffusion, which could eventually consume all the nickel and allow the fuel to come in direct contact with the coolant
- 2) Progressively increasing fuel growth and/or swelling
- 3) A progressive increase in the decomposed coolant surface films.

With respect to the first limitation mentioned, the outer fuel cylinder's outer diameter increase is the critical dimension. Figure 13 shows a plot of the change in the outer diameters along the outer fuel cylinders from the two fuel elements vs their respective fuel burnups. A linear least squares fit of this data shows a 0.20% increase in outer diameter per 4.65×10^{19} fissions/cc

(1000 Mwd/MTU) and a standard deviation of 0.09%. If this linear behavior continues, firm contact between the fuel cylinder cladding and the outer process tube would be expected at a burnup of $2.8 \times 10^{20} \pm 0.9 \times 10^{20}$ fissions/cc (6000 $\pm$ 1900 Mwd/MTU). The bulk of the data plotted in this figure were obtained from clad fuel cylinder measurements, but data from measurement of the bare cylinders are also included. Figures 9 and 10 show that a very good correlation does exist between the clad and bare fuel cylinder outer diameter changes.

In regard to the limitation of the operation of a fuel element caused by a substantial reduction in coolant flow area as a result of fuel growth, an interesting but, as of yet, unexplained phenomenon was observed. The fuel cylinder inner diameter increases were generally larger than the outer diameter increases, such that the volume increase of the fuel cylinders was substantially less than would be expected by studying only the outer diameter changes. As a result, the total decrease in the cross-sectional flow area was very small. The most severe conditions observed in the P-1091 fuel element could account for only approximately a 4% decrease in the cross-sectional flow area.

B. NICKEL DIFFUSION BARRIER

The primary purpose of the nickel plating on the bare fuel cylinders was to act as a diffusion barrier between the uranium alloy fuel and the aluminum cladding. The diffusion rate of uranium in aluminum is relatively high; and, had interdiffusion of these species taken place, the uranium and fission products would have quickly migrated into the main coolant stream. One of the primary advantages of an organic-cooled reactor is the very low induced radioactivity in the coolant, which significantly reduces the machinery vault shielding requirements and allows easy access for maintenance. Therefore, if fuel and fission products were introduced into the primary coolant, this advantage would be lost. Under normal conditions, in the absence of irradiation, the thickness of the nickel deposited would have been more than adequate.⁽²⁾ However, there was some concern about the possibility of a greatly accelerated interdiffusion during reactor operation. Examination of the metallographic specimens did not reveal excessive nickel consumption,

which might have been caused by irradiation-induced acceleration of the diffusion process. The thickness of unreacted nickel in all cases was >0.0008 in., while the minimum thickness of the nickel film was 0.0010 in. after fabrication. A more quantitative statement concerning the amount of interdiffusion one would expect in the absence of the irradiation field was not made, since neither of the two elements examined were instrumented for direct fuel temperature measurements. As a result, the exact temperature at any specific position could only be estimated.

C. FILM DEPOSITS

The examination of the first two fuel elements removed from PNPf Core I has shown that the continuous maintenance of the coolant quality resulted in very low film formation rates, an average of 0.00011 in. per operating year for Fuel Element P-1071, and 0.00058 in. per operating year for Fuel Element P-1091. Neither the film deposits nor the fuel growth were sufficient to reduce the flow area enough to account for the observed increase in the outlet coolant temperature of the P-1091 fuel element. The reason for the observed coolant outlet temperature increase (coolant flow reduction) is believed to be due primarily to a substantial increase in surface roughness, associated with the film deposits on the heat transfer surfaces. Figure 7 shows closeups of the surface of the aluminum cladding, and illustrates that even the very light film deposits possessed very rough surfaces.

The thickness of the film deposits was found to be dependent primarily on the temperature and/or neutron flux level and the rate of coolant flow. Since, in the Piqua reactor, the coolant flow is downward and the control rods enter from the top of the core, both the maximum temperature and maximum neutron flux levels occur at approximately the same vertical position, slightly below the axial centerline of the core. Therefore, it was not possible to distinguish between the effects of temperature and radiation flux on the formation rates of films in these fuel elements. However, experience has shown that the radiolytic decomposition rates are greater than the pyrolytic decomposition rates, for the organic coolants which have been used to date.⁽³⁾ The effect of coolant velocity can be seen quite dramatically in two specific cases. First, on Fuel Element P-1071, the thickness of the surface film on the

outside of the outer process tube was ~5 to 10 times greater than the films observed on the heat transfer surfaces. The velocity of the coolant is several orders of magnitude greater through the fuel element than outside the process tube, where it serves only as a moderator. The second case is the film formations inside of the inner process tube and the valve assembly below it. When the fuel element is in its normal position in the core, the ball plunger in the valve body is forced up into the valve body. In this closed position, the only coolant flow through the inner process tube is through the 7/64 in. drainage hole in the side of the valve body and around the snug-fitting ball plunger. This provides for $< 0.25\%$ of the total flow through the fuel element. In addition to this, the flow through the volume below the drain hole in the side of the valve body and above the ball on the plunger was less than one fifth of the flow through the inner process tube. This small volume of extremely low flow would be highly susceptible to coolant decomposition. Such decomposition could nucleate on any small particle in the coolant which could not pass out of this space. The process of decomposition of the coolant would then progress throughout the small volume, and eventually work its way up into the inner process tube. The condition of the material found in these positions on the two fuel elements examined seem to support this mechanism, since the material became progressively more decomposed and coke-like toward the bottom of the inner process tube and into the valve body.

VI. CONCLUSIONS

The major conclusions drawn from the examinations of the first two fuel elements removed from Piqua Core I are summarized as follows:

- 1) The overall general condition of both fuel elements was excellent.
- 2) Fuel dimensional increases were small, with the change of the outer diameter of the outer fuel cylinders being $0.20 \pm 0.09\%$ increase per 4.65×10^{19} fissions/cc (1000 Mwd/MTU). Assuming fuel dimensional changes continue to be linear with burnup, firm contact between the fuel cylinder cladding and the outer process tube would be expected at a burnup of $2.8 \times 10^{20} \pm 0.9 \times 10^{20}$ fissions/cc (6000 ± 1900 Mwd/MTU).
- 3) The nickel diffusion barrier between the fuel and the cladding did not show a significant increase in its interdiffusion rates as a result of irradiation, and it performed successfully as a diffusion barrier.
- 4) No fuel-cladding separation was found in either fuel element.
- 5) Surface films on the heat transfer surfaces were very thin, showing average film formation rates of 0.00011 and 0.00058 in. per operating year for Fuel Elements P-1071 and P-1091 respectively.
- 6) The accelerated coolant decomposition observed in the inner process tubes and valve body assemblies does not seem to present immediate serious problems which would affect the normal operation of the fuel elements. Careful observation should be made in any of the subsequent fuel element examinations, to assure that serious problems do not develop.
- 7) The progressive increase in coolant outlet temperature for the P-1091 fuel element is believed to have been caused by a substantial increase in surface roughness, associated with the surface films that were deposited on the heat transfer surfaces.

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