Conf-9408206--

PNL-SA-21636

SIMULATION OF NPLWR ENVIRONMENT IN THE ATR LOOP-1 TEST

G. W. Hollenberg
R. S. McKee
R. R. O'Connor[a]
J. R. Thielges[b]
D. R. Spencer[c]

August 1994

Presented at the
Sixth International Symposium on Environmental
Degradation of Material in Nuclear Power System
August 1-5, 1994
San Diego, California

Prepared for
the U.S. Department of Energy
under Contract DE-AC06-76RLO 1830

Pacific Northwest Laboratory
Richland, Washington 99352

(a) EG&G, Idaho Falls, Idaho
(b) Westinghouse Hanford Company, Richland, Washington
(c) Westinghouse Advanced Energy Systems, Madison, Pennsylvania

MASTER

DISTRIBUTION RESTRICTED TO U.S. ONLY

DC

~~DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED~~

SIMULATION OF NPLWR ENVIRONMENT IN THE ATR LOOP-1 TEST

G.W. Hollenberg[1], R.R. O'Connor[2]
R.S. McKee[1], J. R. Thielges[3] and D. R. Spencer[4]

[1] Pacific Northwest Laboratory, Richland, WA 99352
[2] EG&G, Idaho Falls, ID 83415
[3] Westinghouse Hanford Co., Richland, WA 99352
[4] Westinghouse Advanced Energy Systems, Madison, PA 155663

Abstract

An irradiation test of high-temperature, light water reactor, tritium targets was conducted in the ATR that simulated the neutronic, thermal-hydraulic and chemical conditions in an NPLWR for the production of tritium. Requirements for water chemistry, temperature, pressure, spectrum and fluence were met for essentially all of the 217 days of irradiation. The loop test facilities in the ATR provide excellent capabilities for conducting long term, well controlled, in situ tests to simulate degradation of the materials, components and systems used in LWR's and should be considered for testing other components.

Introduction

The New Production Light Water Reactor (NPLWR) concept utilizes high-temperature tritium target elements for production and containment of tritium. The critical issues to be resolved in this test were 1) the tritium production and retention characteristics of the target rod and 2) the general stability of the target rods in a reactor environment. This paper describes the desired reactor environment and the experiment capability of an ATR loop facility to maintain that environment during irradiation. The results addressing the tritium retention and irradiation stability issues will be discussed in a later paper.

Test Description

The Advanced Test Reactor (ATR) at INEL contained 9 individual irradiation loops in a 3X3 array within a water-cooled core. The 4 foot long fuel plates in the core are curved into a serpentine pattern such that they surrounded the central and the four diagonal loops, i.e. the loops at the corners of the 3X3 array. Beryllium reflector/hafnium absorber drums coupled with shim rods individually control the local power levels in each of the diagonal loops. The test volume of this loop was completely separated from the reactor coolant system by a Large-In-Pile-Tubes(LIPT), which is shown in Figure 1. For the Loop-1 test the pressure inside the tube was increase to 2150 psi by the pressurizer system in order to accommodate the prescribed operating temperature of slightly over 300°C. The coolant was routed out of the reactor and into a set of eight circulation pumps which fed a set of heaters before returning it to the LIPT. A small fraction of the coolant(1%) was continuously routed to exchange columns where two types of ion exchange columns were available: one to control the pH of the coolant and a second to stabilize the lithium and boron levels in the coolant. Sampling ports provided the capability to extract aqueous specimens on a periodic basis for subsequent analysis. In addition, aliquots of hydrazine or other chemicals could be injected, if necessary, by this route. To assure that localized boiling did not take place in the loop, the pressurizer provided constant over pressure and, in addition, provided a plenum volume for expansion of the coolant. A "de-gasser" line was continually operated to provide good mixing in the pressurizer. During operation water losses associated with leaks in loop components reduced the level of coolant in the pressurizer. At prespecified coolant levels, the Makeup Water System was activated to inject metered quantities of premixed coolant into the system with the appropriate concentrations of boron and pH for replenishment as needed.

The closure plug(See Figure 1), when opened, permits the insertion and removal of test assemblies into the LIPT. The Loop-1 test assembly which is shown in cross section in Figure 2 consists of a central instrument rod surrounded by five foot long target rods positioned by sections of an LWR grid spacers at 5 axial positions. The target rods were surrounded by a hafnium shroud to tailor the flux spectrum. The instrument rod provided thermocouple access to several axial locations within and below the core. The wall thickness of the hafnium shroud was precisely set in order to achieve the desired neutron spectrum. Supplying adequate cooling water to the hafnium shroud was essential for temperature control.

The high-temperature LWR target rods were composed of four primary components:inner liner, target material, getter, and barrier coated cladding. The target material was thinned-walled, annular, $LiAlO_2$ ceramic pellets which generated tritium by neutron captures by the ^6Li atoms. Some of the tritium atoms diffused out of the $LiAlO_2$ ceramic and entered the plenum as either T_2O or T_2. The inner liner of Zircaloy provides a method for reducing all oxidized tritium to the elemental form. Tritium in the elemental form was readily gettered by a nickel-plated zirconium getter that surrounded the $LiAlO_2$ ceramic pellets. Finally, at low internal tritium partial pressures the aluminized coatings on the inner and outer surfaces of the stainless steel cladding provided a barrier to the loss of tritium from the target rod to the coolant.

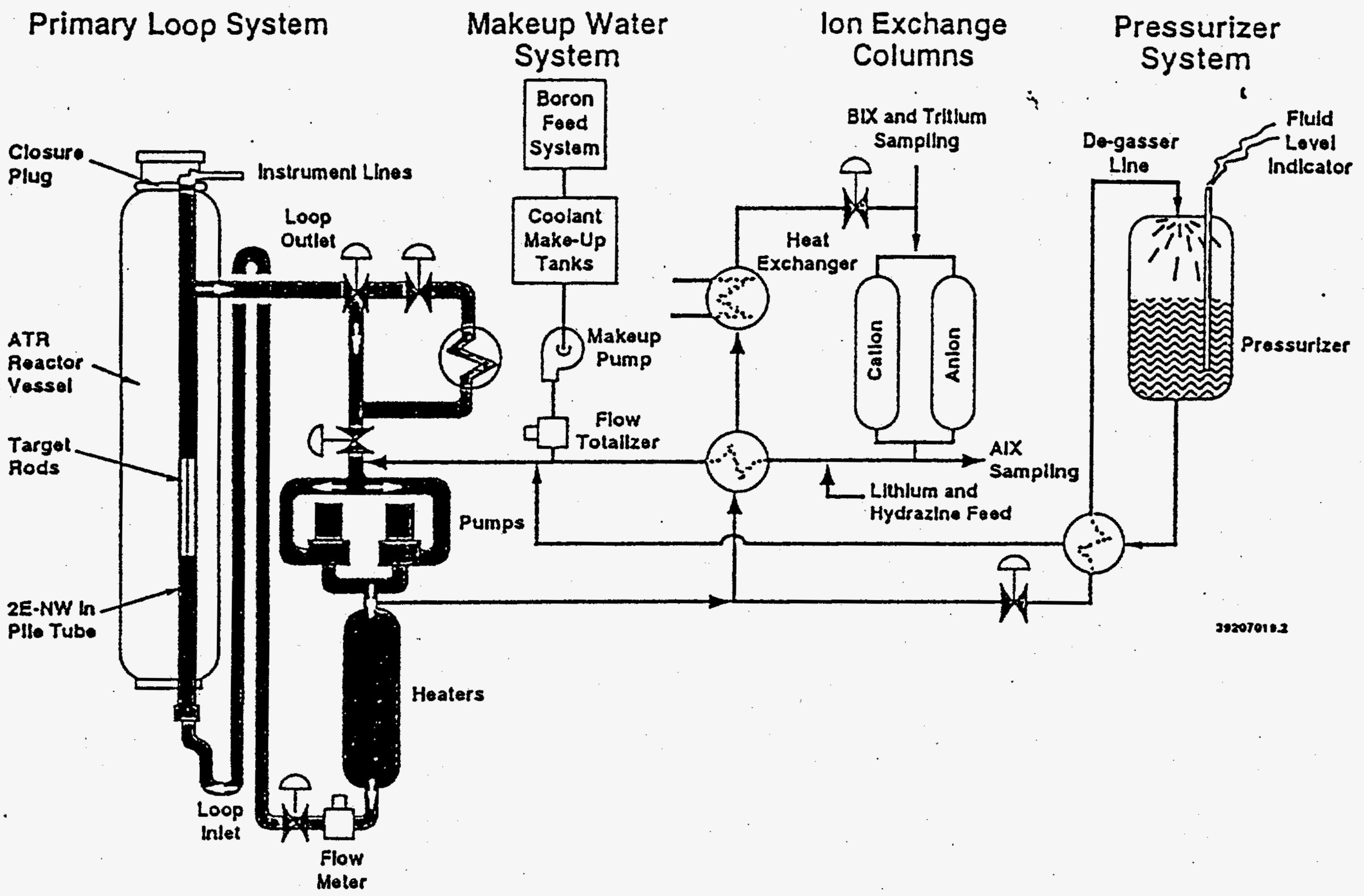

Figure 1. Schematic Diagram of 2E-NW Loop Including the Primary Loop System, Ion Exchange Columns, and the Pressurizer System.

Table I
Comparison of Design Goals for Loop-1 Test with Average, Maximum and Minimum Values Measured during the 217 Days of Irradiation.

Parameter	(units)	Design Goal	Average/1σ	Max/Min
Power	(MW)	23.0±3	22.95±0.16	23.4/20.0
Temperature	(°C)	332.2+0-13.8	318.3±1.3	320.3/316.2**
Pressure of Coolant	(psi)	2150±25	2145±7.7	2197/2116
Total Flow	(gpm)		160.7±2.3	172.4/153.2
pH	(@25°C)	6.65±0.2	6.75±0.08	7.0/5.8
Boron	(ppm)	200±20	203.9±5.9	220/191
Li	(ppm)		0.52±0.08	0.7/0.2
O_2	(ppb)	<15***	12.5±5.1	30.0/5.0
F + Cl	(ppb)	<100	19.5±16.3	93.0/0.0
NO_3	(ppb)		14.7±14.8	55.0/0.0
SO_4	(ppb)		11.8±13.7	37.0/0.0
Al	(ppb)	<50	12.0±3.9	23.0/5.0
Filterable Solids	(ppb)	3	13.3±6.9	50.0/12.0
Conductivity	(μMho/cm)	<10	5.4±0.5	7.7/1.7
Gas Total	(cm^3/Kg)	<140	28.2±8.3	56.6/1.3
H_2	(cm^3/Kg)	30±20	17.3±6.0	46.0/6.0

* Startup and Shutdown power levels not included when power was less than 20.0 MW

** Measured values extrapolated to represent mean cladding temperature axially.

*** 140 ppb of oxygen allowed at startup

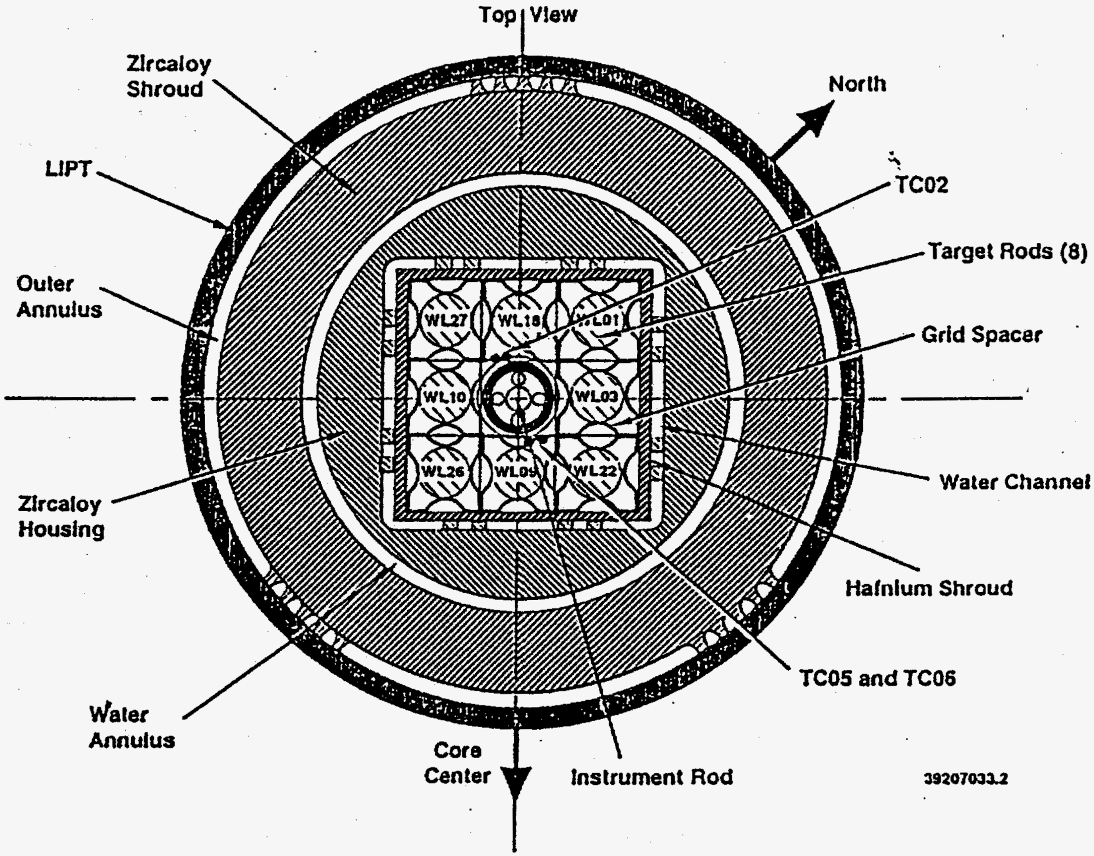

Figure 2. Diametral Cross-Section of the Loop-1 Test Assembly in the Core Region. Note the Identification of the Individual Target Rods.

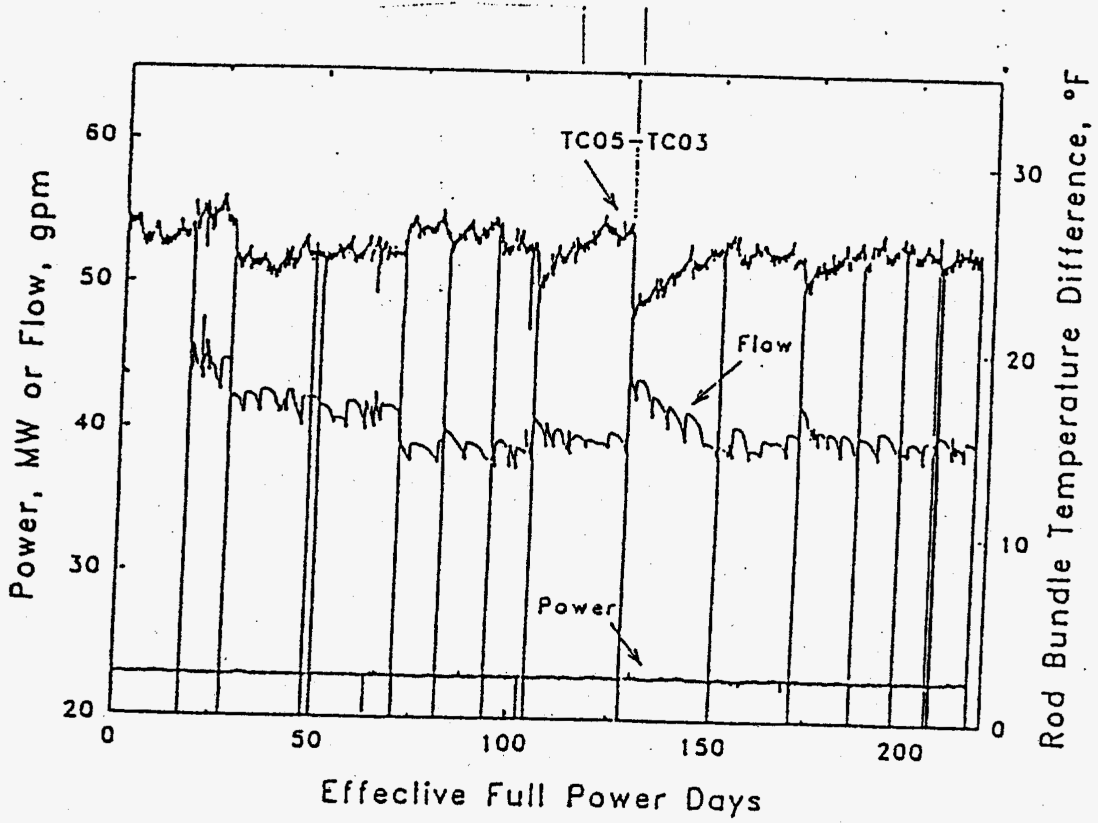

Figure 3. Comparison of the Loop Power, Flow and Rod Bundle Temperature Differences during the Loop-1 Test.

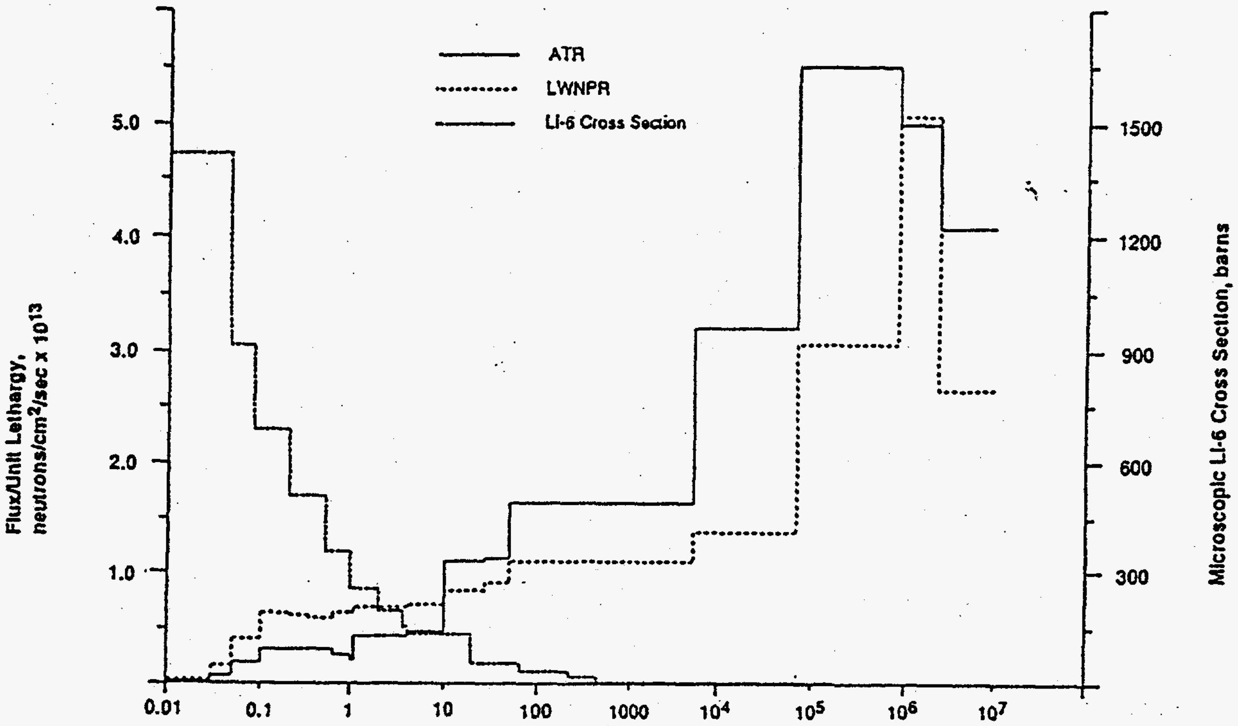

Figure 4. Comparison of the Neutron Spectrum in the Loop-1 Target Rods with Spectral Tailoring with the Spectrum from an LWR used for Tritium Production(LWNPR). The Neutron Cross Section is High at Low neutron energies but the Spectral Tailoring Reduced the Number of Thermal and Epithermal Neutrons at Low Neutron Energies.

Tritium permeation through the cladding was a very small fraction of that generated(0.0075%).

Monitored Test Environment

Operation of the loop test can be separated into two activities: 1) Loop Power and Temperature control and 2)water chemistry control.

Loop Power and Temperature Control

The control of local power in this loop of the ATR reactor was very precise as seen in Figure 3 which permitted highly accurate temperature control. The positioning of the rotating drums is controlled by an algorithm based upon reactor power as determined by overall heating in the entire reactor and local perturbations in flux determined by local flux monitors. Through the 217 days of irradiation the reactor was scrammed or shut off only 15 times. Except for a few hours of slightly reduced power, the Loop-1 experiment ran for 217 days at an average power of 22.95±0.16 MW including the time spent ramping up from 20 MW on startups.

The average temperature of the target rods was dependent upon two independently controlled parameters, the inlet temperature of the coolant and the temperature difference across the rods. The temperature difference across the core, i.e TC05-TC03, varied more than the reactor power, mainly because of variations in the coolant flow as seen in Figure 3. In Table I, the total flow in the test assembly possessed a standard deviation of only 1.5%. A slight recalibration of the flow controller altered the measured difference several degrees at approximately 125 days into the irradiation. Because of this larger variation in flow the average cladding temperature in Table I as extrapolated from the outlet temperature(318.3°C) possessed a standard deviation of ±1.3°C.

Spectral tailoring in the Loop-1 experiment was essential in order to obtain the correct lithium-6 reaction rate in the target rods. Many of the performance parameters were monitored with respect to lithium-6 reaction rates. The cross section of the Lithium-6 isotope is highly dependent upon spectrum as shown in Figure 4. Lower energy neutrons in the unperturbed ATR spectrum would have caused an excessively high reaction rate, over-pressurization of the rods, premature burnout of the lithium-6, and gradients in heating and tritium production due to self-shielding. A zero-power critical facility associated with ATR, along with analytical extrapolations, were used to determine nuclear reactivity of the assembly and to correctly size the thickness of the hafnium shroud so that a flux and spectrum near those of a tritium-producing LWR could be obtained. The unperturbed spectrum in the ATR was expected to be a thermal flux of $4.3X10^{14}$ $n/cm^2/s$ and a fast flux of $3.8X10^{14}$ $n/cm^2/s(E>.1Mev)$. With the hafnium shroud in place the predicted thermal flux was reduced to $0.1\ X10^{14} n/cm^2/s$ and fast flux was

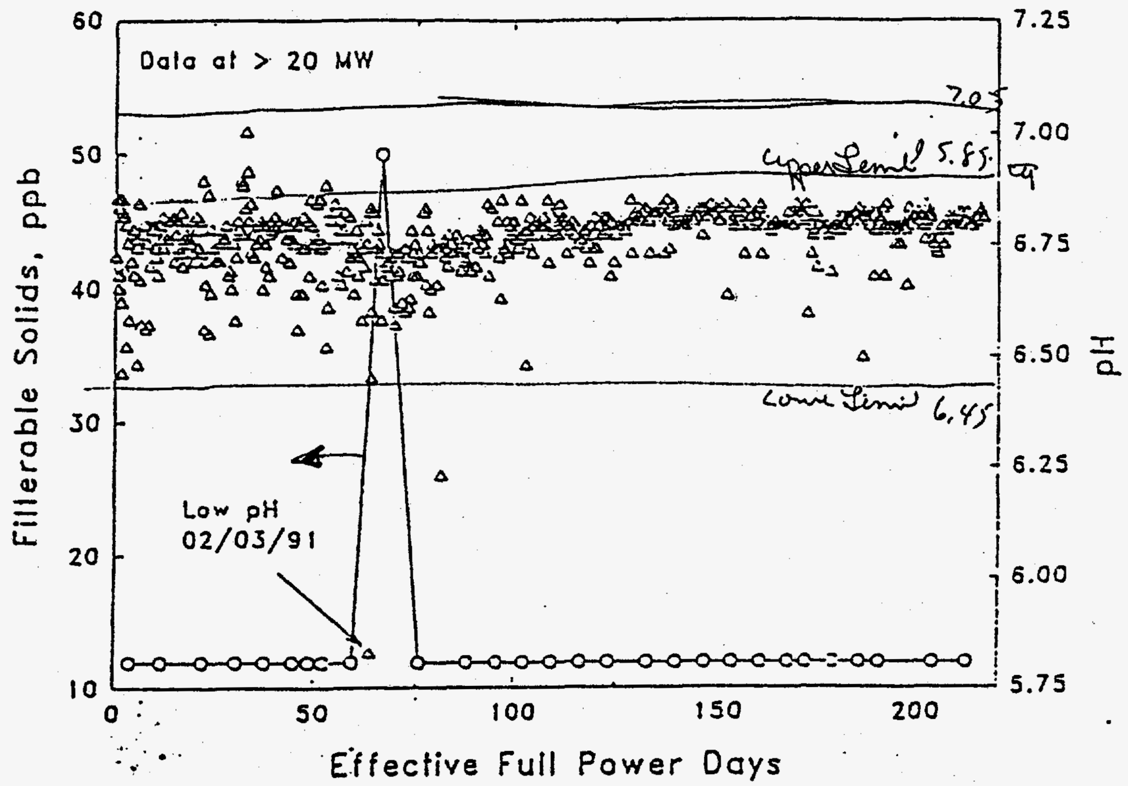

Figure 5. Comparison of the Filterable Solids and pH during Loop-1 Irradiation. Note the Large Increase in Filterable Solids immediately after the Low pH Value was Observed.

maintained at greater than 1.5 $X10^{14}n/cm^2/s$. It is seen in Figure 4 that the 0.084 inch thick hafnium shroud did reduce the flux at the lower energy levels to more desirable levels. The predicted variation in burnup between target rods was from 11.7 to 12.8% ^{6}Li with an average burnup of 12.3% ^{6}Li. The predicted peak-to-average ratio for burnup along the 4 foot target rod was 1.46. The predicted, overall tritium production in the eight rods was 12473 Ci.

Chemistry control

Control of coolant chemistry was one of the major advantages of testing in an ATR loop facility but was also the most labor intensive part of the testing. During operation it was necessary to take water samples at 8 hour intervals (i.e. over 700 measurements) for boron, pH and conductivity measurements. These labor-intensive measurements were necessarily completed in a rapid manner so that any changes could be affected prior to the next measurement. For example, maintaining pH to within 6.45 to 6.85 at 25°C was considered important because of the corrosive effects of high pH values on the aluminide coating on the target rod and the deleterious effects of low pH values on the transport of radionuclides on reactor operations. The Loop-1 test provided an excellent example of this latter point when on 2/3/91 the pH of the loop drifted below the lower limit to a value of 5.83 for a period of less than 5 hours when one of the ion exchange columns was valved in too long(See Figure 5). As a response to the decreased pH, the filterable solids in the loop coolant increased rapidly to 50 ppb and then returned to its lower level, i.e. a "Crud Burst". The location of these particles, i.e. ion exchange columns, low flow regions in the loop, etc., prior to being deflocculated and dispersed into the system was not identified. In an actual reactor the filterable solids would contain radioactive corrosion products and in a pH excursion, like this one, they would be redeposited in locations where they could increase the radiation dose to workers. The excellent control of pH during the rest of the experiment demonstrated that filterable solids can be minimized by proper pH control.

The control of pH was accomplished by several independent chemical parameters of the coolant: boron content, lithium content and other additives. In an actual LWR, boron (a burnable poison) and its associated reactivity are reduced in the coolant during a cycle as the reactivity of the fuel decreases due to its own burnout. The range in boron content could be from 2000 ppm to 0 ppm. A value of 200 ppm was selected as representative for this irradiation and was maintained at that constant value. The lithium content of approximately 0.5% yielded the correct pH range. Stability of these components was maintained by injection of carefully premixed makeup water as water leaked from the loop. Many of the other chemical parameters were measured on samples taken at intervals of once per day.

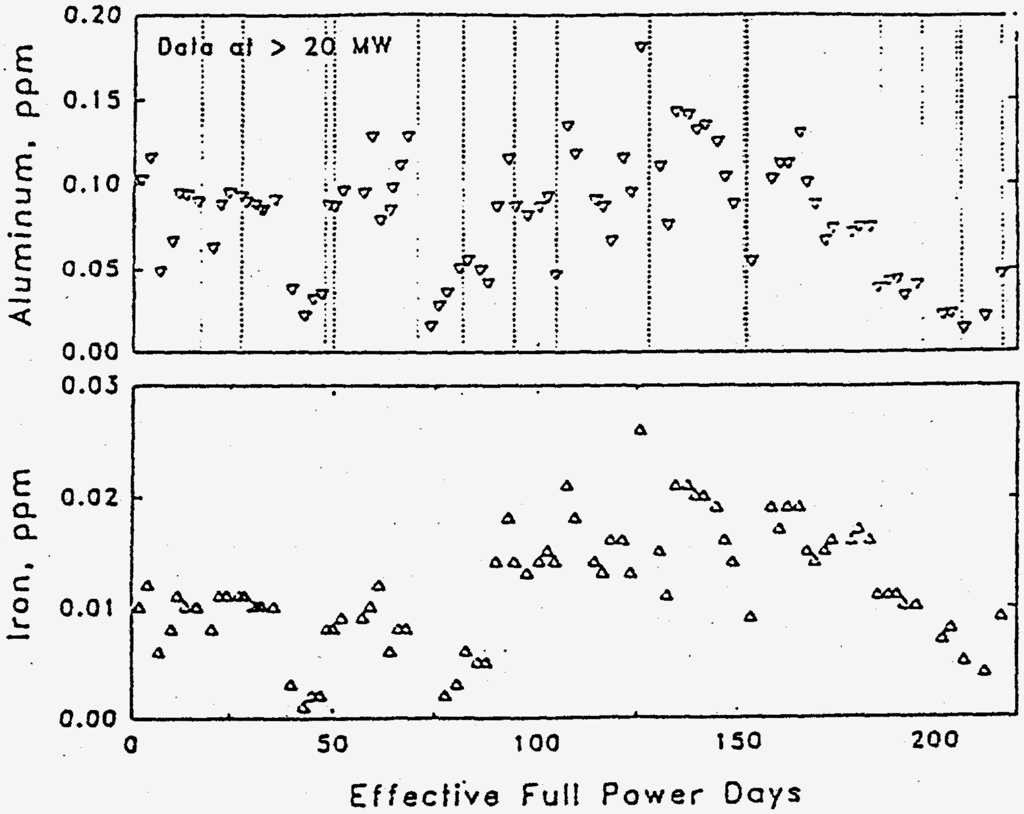

Figure 6. Measured Aluminum and Iron Concentrations in the Doolant During the Loop-1 Irradiation.

The oxygen content of the coolant is important in controlling the corrosion of components throughout the system. Oxygen was maintained at low levels by elevating the hydrogen content in the coolant via injections of hydrazine hydrate($N_2H_4 \cdot nH_2O$). During the start up of the reactor the oxygen content was sometimes higher than 15 ppb. However, the target rods were also cooler so corrosion was limited.

In Figure 6 the concentration of iron and aluminum were found to be essentially proportional to each other and many other elements during the irradiation. Corrosion of these elements in the target rods or loop components was anticipated. However, the effects of the system's ion exchange columns on reducing the concentration in solution contributed to a more complex relationship. Analysis of this data indicated that instead of a dependence on factors which influence corrosion via chemical analysis, the concentration of aluminum, iron, and other impurities in the coolant(Ca, Mg, Na, Fl, Cl, etc.) were more dependent upon the leak rate and hence the rate of injection of clean water into the loop system. Hence, it was not possible to monitor corrosion in the loop due to the presence of the ion exchange columns.

The loop test assembly was removed from the core after irradiation and is now awaiting funding availability for post irradiation examination. Tritium permeation into the coolant during irradiation was determined during the irradiation but will be reported in detail at a later time. The low tritium permeation observed during the Loop-1 experiment provided confidence that the target rod design was successful.

Conclusions

The Loop-1 irradiation experiment was successful in providing a test bed for testing tritium target rods designed for use in a tritium-producing LWR. The ATR spectrum and flux were successfully tailored to provide a desired reaction rate and spectrum-averaged cross section. The thermal control provided by the Loop-1 components allowed for precise cladding temperatures to be achieved during irradiation. The intense efforts of chemists permitted the control of many chemistry parameters in the coolant throughout the irradiation.

Acknowledgements

An experiment as large and complex as the Loop-1 test requires the support of many contributors. The design, chemistry, computer and operations supported by Don Lanning, John Matusz, Phil Erickson, Nancy Wildung, Tim Comstock, George Simmons and Guy Johnson were especially noteworthy. The management efforts of Larry Garrelts, Mike Travis, Jerry Ethridge, Max Feshley and Ray Puigh were necessary in order to conduct a test of this magnitude. Funding was made

DO NOT TYPE ON THIS LINE

First line of text (other than title page)

First line of title; all caps and center

Second line of title

Authors' name, center

Authors' affiliations, center

available under U.S. Department of Energy Contract DE-AC06-76RLO 1830.

DO NOT TYPE IN BOX

rev. 3/83