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PROGRESS REPORT ON THE
ACCELERATOR PRODUCTION OF TRITIUM
MATERIALS IRRADIATION PROGRAM

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Abstract

The Accelerator Production of Tritium (APT) project is developing an accelerator and a spallation neutron source capable of producing tritium through neutron capture on He-3. A high atomic weight target is used to produce neutrons that are then multiplied and moderated in a blanket prior to capture.

Materials used in the target and blanket region of an APT facility will be subjected to several different and mixed particle radiation environments; high energy protons (1-2 GeV), protons in the 20 MeV range, high energy neutrons, and low energy neutrons, depending on position in the target and blanket. Flux levels exceed $10^{14}/cm^2s$ in some areas.

The APT project is sponsoring an irradiation damage effects program that will generate the first data-base for materials exposed to high energy particles typical of spallation neutron sources. The program includes a number of candidate materials in small specimen and model component form and uses the Los Alamos Spallation Radiation Effects Facility (LASREF) at the 800 MeV, Los Alamos Neutron Science Center (LANSCE) accelerator.

Introduction

Concept

The APT program is designing an accelerator capable of producing a proton current of around 100 mA in the 1-2 GeV energy range; optimization studies considering production needs, plant cost and operations cost will determine the energy eventually chosen. The protons are directed to a target made of a high atomic number material such as tungsten. Spallation reactions produce neutrons as well as protons and other particles. The neutrons produced have an energy spectrum that extends to the proton source energy.

Target and Blanket

The primary neutron source is surrounded by a blanket made of lead which serves both as a neutron multiplier and moderator. In the blanket, neutrons are captured in He-3 producing on evaporation tritium and a proton. A back-up concept uses an alloy of Al 3% Li enriched in Li-6 producing after capture and evaporation tritium and He-4. In the gas system an on-line separation plant continuously removes tritium from a flowing system; in the Li system the alloy is irradiated for a period of time, removed from the facility and processed in another facility. Figure 1 shows a schematic cross section of the He-3 target and blanket assembly.

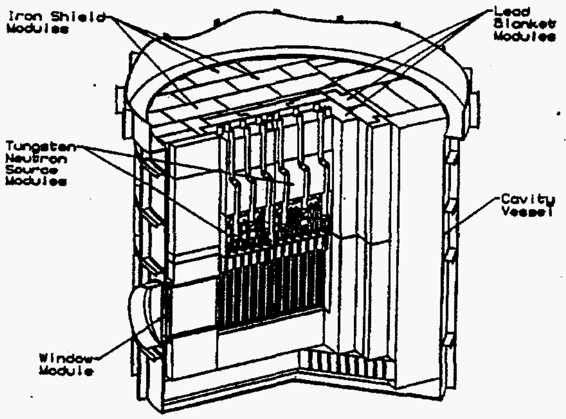

Figure 1: Schematic cross section of an APT target and blanket assembly. one half of the blanket assembly has been removed for clarity.

Radiation Environment in the APT Target and Blanket

Referring again to Figure 1, note that the proton beam enters the target and blanket assembly through the window module before interacting with the tungsten neutron source module. The primary proton beam is restricted in size and does not directly impinge on the decoupler or other parts of the blanket. The target and blanket assembly is exposed to the primary proton beam at an energy of 1-2 GeV. For a beam expanded to a size of 16 by 160 cm, the proton fluence in nine months of operation will be about 5.8×10^{21} protons/cm^2 for a 100 mA current and 7.7×10^{21} protons/cm^2 for 134 mA; these are the two likely currents in an operating plant. The leading part of the tungsten neutron source assembly will also be exposed to the primary proton beam; the energy of the protons will decrease as a function of length while passing through the neutron source assembly. A neutron flux will also be generated as the beam passes through the neutron source assembly resulting in a mixed proton and neutron flux. The proton and neutron energy spectra will also change as a function of distance into the target. The decoupler region will be exposed to high energy neutrons with energies up to a few hundred MeV and protons produced by spallation reactions in the target with energies in the several tens of MeV range. The remainder of the target/blanket assembly including the lead modules, reflector modules, and shielding will be exposed to a mixed-spectrum of high and low energy neutrons that varies as a function of position. Calculations, using high and low energy transport computer codes including the Los Alamos High Energy Transport code (LAHET) in the LAHET Code System (LCS), predicting the flux-spectra throughout the target and blanket are performed along with optimization studies of the configuration. Proton fluxes above $10^{14}/cm^2s$ are expected.

Neutron fluxes approaching $10^{15}/cm^2s$ are also expected in some regions.

Radiation Environment Considerations

Development of radiation-resistant and radiation-tolerant materials for use in fission reactors has received substantial attention over the past 50 years. More recently, the fusion reactor initiatives have required additional materials radiation damage effects studies since here the neutron energy is higher, 14 MeV opposed to an average of about 1 MeV for a typical power reactor. The information learned in these programs is being applied to the design of the APT.

As mentioned above, the target and blanket components are exposed to particle radiation with energy up to 2.0 GeV (current design is at 1.7 GeV). Only a limited data base is available for materials applications in this environment. Because of experience at accelerators such as at the Los Alamos Meson Physics Facility, now Los Alamos Neutron Scattering Center (LANSCE), phenomenological evidence indicating good material performance of a limited number of structural materials and target materials for adequate operating times is available. Determination of design data such as strength and ductility has been limited to very low dose experiments.

As the particle energy increases to above 200 MeV or so a major new variable becomes important; copious transmutation products are generated and enter metal alloy systems as impurities. For example, helium atoms produced in a metal lattice have been found, by the fusion reactor community, to have a major effect on materials properties. Fourteen MeV neutrons produce helium through an (n,α) reaction while 200 - 1300 MeV protons produce about 100 times more helium per particle through spallation reactions. A number of calculations of expected radiation damage parameters in spallation neutron source environments have been done [1-7]. These reports also compare the spallation environment to fission reactor environments where extensive data on radiation damage effects has been generated.

Corrosion-related phenomena have been studied at fission reactor installations and difficulties have largely been solved. Less is known about cooling systems at accelerators; difficulties have been encountered but a basic understanding of the governing conditions is not known. A synergism between charged particle interactions at metal surfaces in contact with water in which substantial radiolysis products are present is expected to be the driving force for corrosion. Details of tests sponsored by the APT program are being reported at this conference [8,9].

Design data on materials performance in prototypical particle radiation environments is essential for proper development of the APT target and blanket. Fortunately the LANSCE accelerator has facilities that allow tests in nearly prototypic environments. At the LANSCE beam stop, the Los Alamos Spallation Radiation Effects Facility (LASREF) is being employed to expose all candidate materials to a particle flux and spectrum that closely matches that expected at APT.

Irradiation Facility

Figure 2 is a schematic of LASREF. Specially designed capsules containing candidate materials or models of prototypic components are placed on the bottom of 3 1/2 m long shields (inserts) where they are exposed to a proton flux, a mixed proton and neutron flux or a neutron flux, depending on where they are located in the facility. Figure 3 shows a close up of the irradiation area at LASREF. Capsule 17A is exposed primarily to the incident proton beam, thus the candidate window materials were placed here. Capsule 18A was constructed to house models of tungsten rod bundles as are being designed for the APT plant. As a function of length through the capsule, the proton energy decreases and a neutron flux develops, also relatively prototypical of the plant. Capsule 18B contains capsules that are not exposed to the direct beam but rather to the mixed neutron and spallation-produced proton flux typical of that present in the APT decoupler region. Capsule 18C contains primarily structural materials that will be exposed also to mixed spectra. There are also capsules and component modules in the region marked "neutron irradiation" in Figure 2. Placement of the specimens was determined through an extensive LCS calculation [10] that modeled the facility and compared the radiation environment with that expected at the plant. To a large measure, the environments for the samples are prototypic in spectrum. The proton current density is also prototypic; neutron fluxes are somewhat lower than expected at APT.

Study Summary

Design data such as strength, ductility and toughness are being determined. Models of the tungsten neutron source rod bundles are being irradiated with 800 MeV protons at prototypic average power levels. Models of blanket modules are being irradiated in a near prototypic neutron environment. Capsules containing He-3 gas are included to determine tritium retention characteristics under irradiation. A closed loop water system will be used to determine water chemistry, corrosion rates of candidate materials, instrumentation capabilities. and corrosion-mitigation methods during irradiation. Samples of weldments and samples used to study stress corrosion cracking susceptibility under irradiation will be studied.

Irradiation Program Outline

Candidate materials for the various components for the APT target and blanket were chosen based on experience at accelerators and fission reactors. Neutronics issues such as neutron absorption cross sections were a strong consideration. A survey of applicable materials has been compiled [11]. Two international conferences on the use of materials at spallation neutron sources have also been held [12,13]. Table 1 summarizes the materials selected for the APT target and blanket.

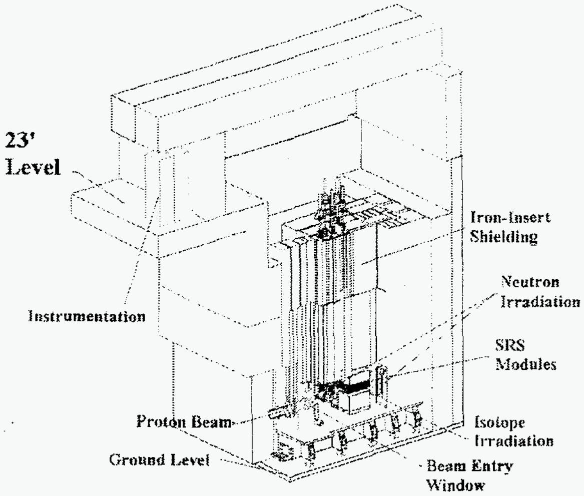

Figure 2. Schematic diagram of the LASREF at LANSCE

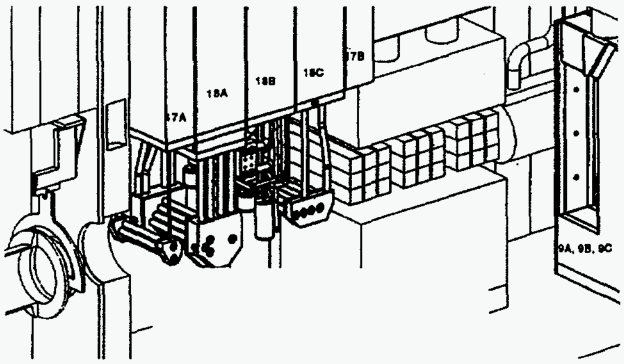

Figure 3. Close-up of the arrangement of capsules used to expose samples and prototype components to the radiation environment at LASREF.

In addition to the materials given in Table 1, the test matrix includes modified austenitic stainless steels, ferritic stainless steels HT-9 and F-82 H and a number of pure metals; the pure metals will be used as a tool to determine the basic aspects of the effect of spectrum on microstructural evolution in an attempt to learn how to better use the extensive fission reactor data base in spallation neutron source applications.

Mechanical properties data are a primary need for design. To obtain these data, samples types as given in Table 2 were included in the test matrix.

In addition to the standard sample types, models of the tungsten rod bundle neutron source were constructed as shown in Figure 4. Nineteen 3 mm diameter rods were arranged on a triangular pitch and placed in a capsule on insert 18A.

Modules made of lead enclosed by aluminum alloy as well as aluminum tubes with He-3 gas and rods of AlLi alloy were

Table 1 Materials Selection of the APT Target and Blanket

Component	Material	Reason	Backup
Beam entry window	Inconel 718	Experience at LANSCE, high temperature strength	316 SS
Target	Tungsten	High neutron production	Tantalum
Target Structure	316 SS	Irradiation performance, neutronics	Inconel 718
Decoupler Structure	Aluminum alloy 6061	Low neutron absorption	Aluminum alloy 5052
Reflector	Lead	Neutron multiplier, low neutron absorption	
Coolant Tubes	Aluminum alloy 6061	Low neutron absorption	Aluminum alloy 5052
He-3 Tubes	Aluminum alloy 6061	Low neutron absorption	Aluminum alloy 5052

Table 2 Specimen Matrix

Type	Purpose
Miniature sheet tensile samples	Strength and ductility Slow strain rate - stress corrosion cracking
Transmission Electron Microscopy discs	Shear strength (microstructure)
Compact tension specimens	Fracture toughness
Bend specimens	Bend strength - Ductile-to-brittle transition - (crack initiation/growth)
Pressurized tubes	Creep
U-bend specimens	Stress corrosion cracking
Weld specimens	Test all expected weld types for strength

Figure 4 Photograph of a tungsten rod bundle during construction

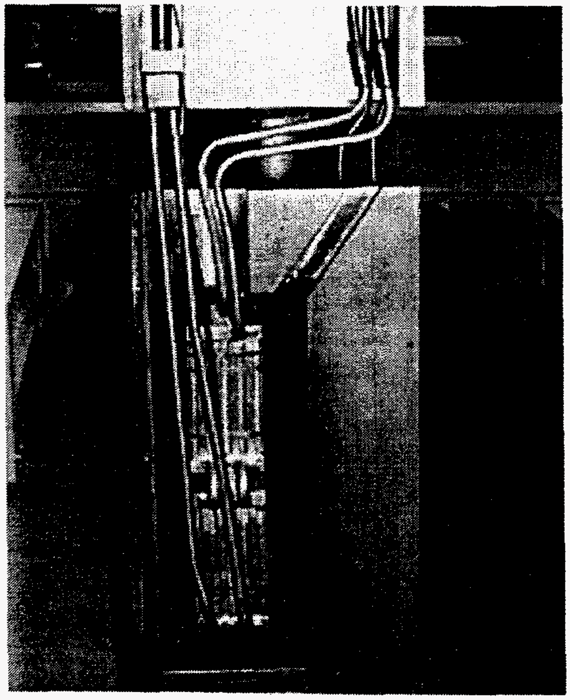

Figure 5. Photograph of a model of the blanket module.

placed in the neutron flux area. This installation can be seen in figure 5. Also included in this irradiation are a number of aluminum tubes filled with He-3 gas. Some of the tubes have been modified by removing the aluminum oxide coating on the tube interior and replacing it with a approximately a micron thickness of Ni. These tubes are further enclosed by another tube, the annulus is filled with water. This arrangement closely models the APT concept. Post-irradiation analysis of these materials will determine where the produced tritium resides.

Irradiation Capsule Development

Capsules for Use in the Proton Beam

The proton beam at LASREF has a gaussian - like space profile with a diameter of approximately $2\sigma = 30$ mm, where σ is the standard deviation. This beam profile has two basic effects on sample placement: 1. to assure reasonable uniformity in dose across a specimen the specimen must be small, and 2. proper placement of specimens presents an opportunity to study the effect of dose in a single capsule. Samples must also be relatively thin, in the 0.25 - 2 mm range to assure that the energy deposited by the 800 MeV proton beam is properly transferred to coolant.

Referring back to Figure 3, it is seen that each capsule is a series of tubes attached at each side by a plenum. Water flows into one plenum, across the tubes that are in the proton beam and out the opposite plenum. Inside the tubes are envelopes containing the specimens. Each specimen type requires a special envelope consisting of a central spacer plate and two cover plates that isolate the specimens from direct contact with the water. The envelope is sealed by welding using special cooling blocks to limit specimen temperature during the sealing process. Several envelopes in a tube can be seen in schematic form in Figure 6. An envelope containing compact tension specimens is shown in the top photograph in Figure 7. The small discs shown are 3 mm in diameter and are each a package of specimens made from Ni, Co, Fe, Cu, Nb, and Al. These foils will be used for dosimetry following the irradiation and will allow determination of achieved fluence and spectrum. Figure 7 (bottom) shows an envelope containing both tensile specimens and bend specimens. Figure 7 shows the envelopes before the final cover has been attached. Each envelope thickness was sized after considering the power deposition and the expected operating temperature at an APT plant. Several thermocouples were placed in the envelopes to measure the achieved temperature during irradiation. As a reference, the envelopes shown are about 159 mm long.

Figure 8 shows the assembled capsule 17A before it was attached to its facility insert. Water flow tests were performed to assure proper flow and pressure drop in each unit before installation. Construction of capsule 18C followed the same procedure as described for capsule 17A. In addition to the specimens envelopes, one tube contained clad 3 mm diameter W rods made in the same manner as those installed on capsule 18A and two tubes contained clad 10 mm diameter rods.

Assembly of the W rod bundles was mentioned above. The capsule containing these units is named 18A. The initial capsule 18A was made up of 8 tungsten rod bundles, two bundles in which the tungsten was in cylinder form, and six tubes, out of the direct proton beam, containing aluminum alloys. The out-of-beam position closely prototypes the

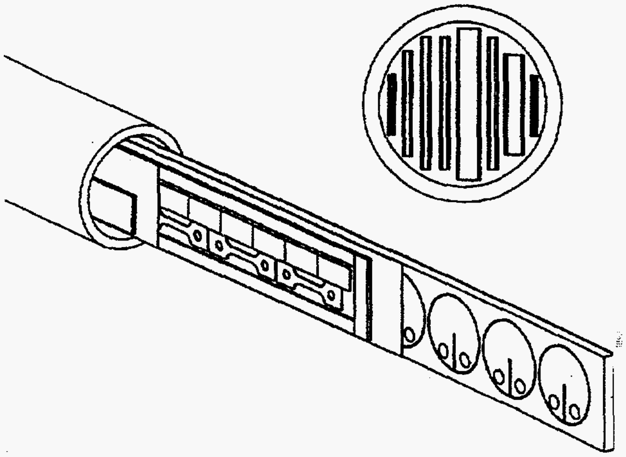

Figure 6. Schematic of samples placed in envelopes and installed in a tube used to direct coolant water across the assemblies.

environment expected in the decoupler region of the APT Target and blanket.

Capsules for Use in the Neutron Environment

Thirteen canisters were built to house standard samples as well as a variety of specimens made from weldments. These canisters were essentially furnaces using a central heater surrounded by specimens. A schematic diagram showing these canisters is seen in Figure 9. Figure 10 shows a canister being loaded with specimens. The temperature of each set of specimens is monitored with a thermocouple and each canister contained a number of activation foil packages. Figure 11 shows Insert 18B which housed 8 of the canisters as well as the He-3 containing capsules and a model of a blanket module. Additional canisters were placed on an insert named 9B which exposed the materials to a spallation neutron flux but without a large proton flux. Insert 9B also housed several specimens used for determining if stress-corrosion cracking would be a concern at an APT plant. In the same area, insert 9A and 9C were fitted with models of the blanket modules. Figure 5 shows the installation of a blanket module on insert 9A and Figure 12 shows the entire insert.

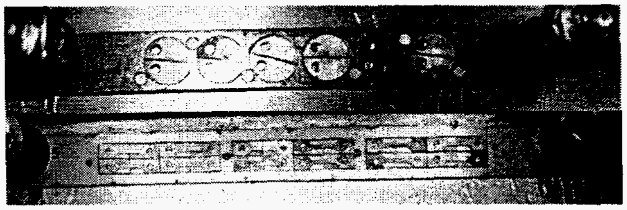

Figure 7. (top) Photograph of an envelope containing compact tension specimens and activation foils. (bottom) Photograph of an envelope containing tensile and bend specimens.

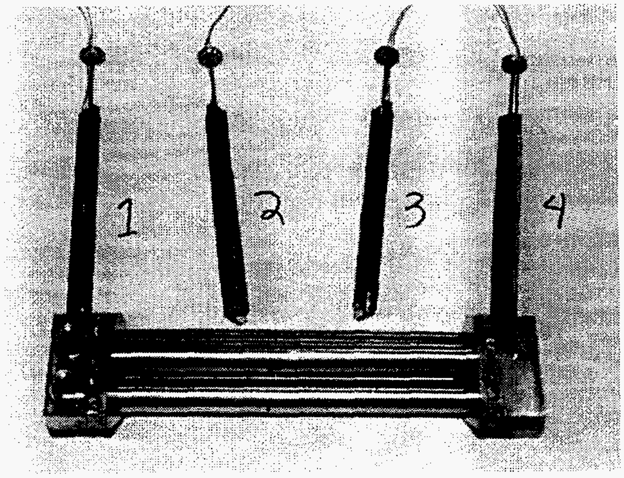

Figure 8. Photograph of capsule 17A before installation on insert 17A.

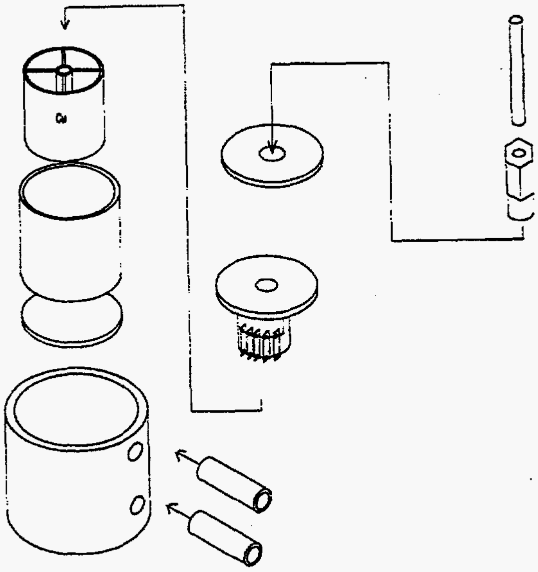

Figure 9 Schematic of a canister used to house specimens in the spallation neutron environment.

Operation Summary

Schedule

Installation of the inserts, capsules and a closed-loop water system for the tungsten rod bundles on Insert 18A was completed in August 1996. The beam was started on August 31 and held at a current of 0.5 mA for two days. The current was gradually increased and the design current of 1 mA was achieved on September 6. After several days of steady operation, several events caused both short and long interruptions. The most significant ones are discussed below.

Figure 10. Photograph of a neutron canister being loaded with specimens.

A water leak in capsule 18A caused termination of the 1996 operations one month earlier than desired. During the two months of operation it is estimated that samples positioned in the center of the beam received a fluence of about 2 x $10^{21}/cm^2$. Work began immediately in November on redesigning and rebuilding capsule 18A.

Water Systems

The pump motor for the closed loop water system serving the tungsten rod bundle capsule failed after only a few days of service. Investigation showed that the controller was supplying a pulsed signal that was being amplified following transmission in a cable longer than normally used. This was corrected, a new motor was installed successfully in spite of the fact that the area was highly radioactive, and the water system operated well for the remainder of the run cycle.

Provision was made to remove samples of the coolant water in the closed loop system. Initial attempts to sample the water resulted in higher-than-expected radiation levels in the LANSCE facility because entrained radioactive gas in the system operating at about 90 pounds/in^2 expanded rapidly and evaporated from the liquid when exposed to only one atmosphere of pressure. This problem was corrected by a procedure that assured that gas evolution during depressurization was contained in the facility air-handling system for radioactive gas.

Water is introduced to the closed loop system at high purity and low conductivity after being treated in an-ion-exchange resin bed. Samples have been taken and analyzed following a series of radiation doses to determine both radioactive and non-radioactive spallation and corrosion products.

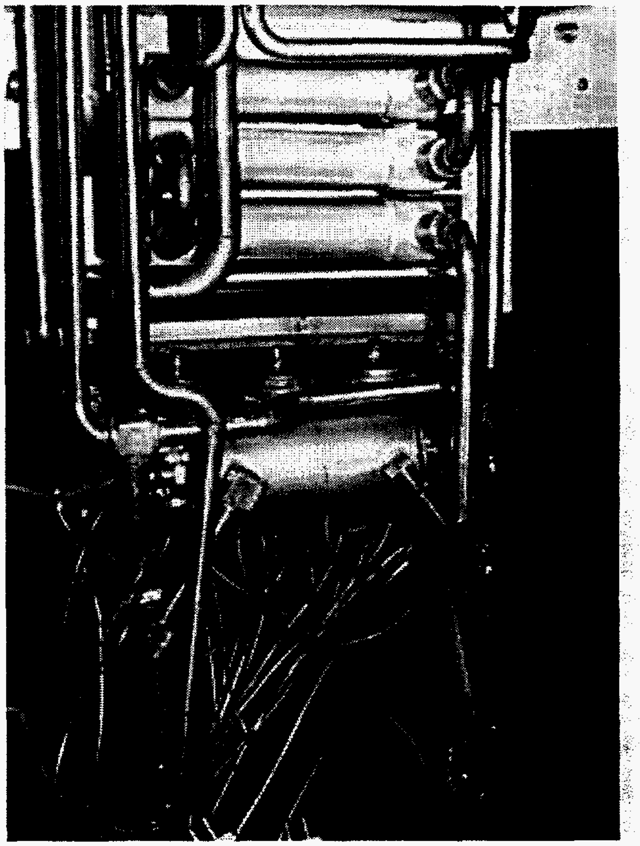

Figure 11. Canisters mounted on insert 18B. Also seen is a housing for the He-3 filled capsules and the blanket models.

Capsule Performance

Measurements of temperature in the proton irradiation capsules using thermocouples placed on prototype samples indicate that design temperatures are being achieved; the energy is supplied only by the proton beam. Temperature readings are recorded in two minute intervals.

In late October 1996, a water leak was discovered in the closed loop water system that serves to cool the tungsten rod bundle capsule (18A). Investigation using diagnostic measurements on the system indicated that the leak was in the capsule within the irradiation area and thus not repairable in the remaining one month of planned LANSCE operation. The irradiation for this cycle was terminated and work began immediately on removing and investigating the capsule. This investigation showed that braze joints used to seal thermocouples that penetrate the capsule plenum had failed. Design and construction of a replacement unit began immediately.

The temperatures of the neutron capsules (furnaces) were set to be prototypic to those generally expected at an operating APT; in the 90 - 150 °C range. Initial operations proceeded as

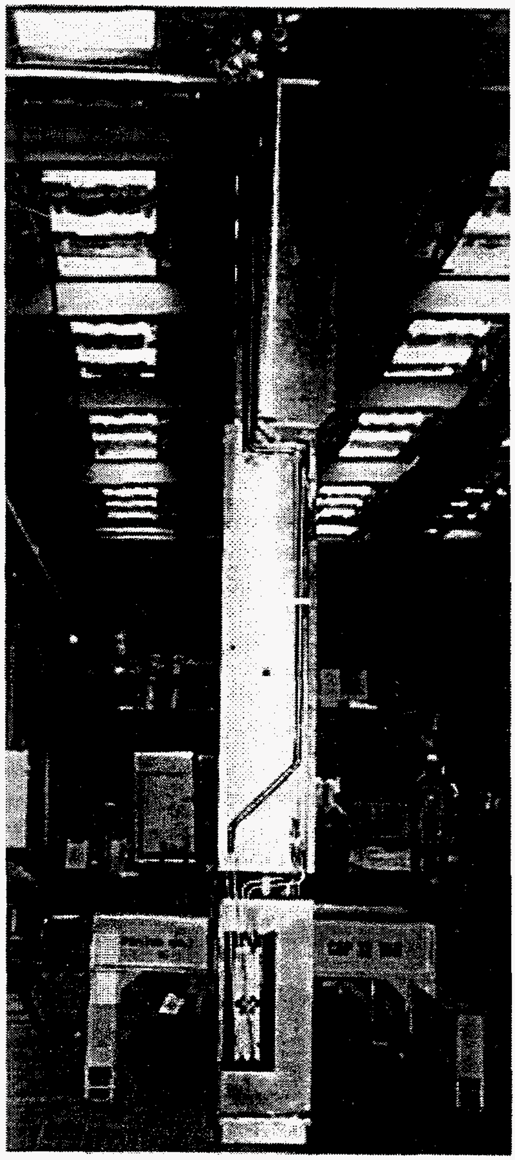

Figure 12. Photograph of insert 9A. The length of the insert is about 3 1/2 m.

planned, however, in time water leaks in a companion facility likely caused the failure of the heater circuits by spraying high velocity water on the current leads. Water leaks were repaired as heaters failed only to occur again with time. Nine of the 13 furnace heaters have failed, fortunately the design is such that heating of the capsule by the radiation source places the samples at a prototypic temperature.

The water circuit serving the capsules on insert 9B also developed a water leak. Investigation showed that the leak was in an inaccessible area. It was also determined that long-term operation with this water leak was not possible Fortunately it was possible to cool these capsules with flowing air; an air leak does not compromise the operations.

Preparation for Additional Operations.

Corrosion-Related Measurements

Development of another closed-loop water system has been completed. This unit will be coupled to a number of capsules that contain U-bend specimens for determining if stress-corrosion cracking occurs in the APT candidate materials in a prototypic environment. Also included in the circuit are a number of probes that will measure corrosion rates during irradiation, some of the probes are directly in the beam, some are in the neutron flux and some are in irradiated water. Weight-loss specimens are also included; these specimens yield an integral of the weigh loss due to corrosion during the entire irradiation period.

The water system is heavily instrumented; measurements of pH, conductivity, hydrogen concentration and system parameters such as flow and pressure are monitored. Provision has been made to allow introduction of chemicals that can change pH as well as hydrogen gas expected to be effective in limiting peroxide formation and thus retarding corrosion. The goal is to determine the water chemistry that produces the minimum corrosion rate for the system. Water samples can also be taken and analyzed to determine dissolved ions and particulates.

Rebuild of Capsule 18A

Capsule 18A is now designed to have no braze joints; thermocouples are placed in thermal wells. The tungsten rods are all clad with a close-fitting stainless steel tube; this decision followed the observation of larger-than-expected tungsten concentrations in the water samples analyzed during the last operation period. The origin of the tungsten is under investigation.

Study of Irradiated Beam Entry Windows Used at LANSCE

Beam-entry windows made from alloy Inconel 718 and used at LANSCE are being studied through a LANSCE-European Spallation Source collaboration at Forschungzentrum-Juelich in Germany. Bend tests and hardness measurements made on samples of a window show that the material has retained strength and ductility at a dose level of approximately 7×10^{21} protons/cm^2. Ductility did decrease as fluence increased. Additional work on these materials is proceeding; finite element stress analysis is necessary to quantify stress and strain from the load-deflection information in a bend test.

Planning

Hot Cell Preparation

Following the irradiation, the capsules will be removed from their inserts and transported to hot cells that are now being prepared with fixtures capable of handling highly radioactive materials and removing the specimens without straining them.

Provisions are also being made to open and study capsule 18a and later, the models of the blanket modules.

Shipping Arrangements

Samples will be packaged and shipped to several laboratories for study. Realizing that shipment of radioactive materials requires a good deal of information and preparation, preparation of documents has already begun.

Schedule

The irradiation of specimens will begin again in March 1997 and continue through July. The capsules will be removed from the inserts by September 1997 and taken to the hot cell facility. Shipments of radioactive samples is expected in December 1997, January 1998. Initial results will be reported in summer 1998.

Organization and Participants

A Materials Working Group has been formed under the APT project structure. This group advises the design teams on materials matters in general and has plans to develop a Materials Handbook or Design Guide containing both published data on non-irradiated material, materials studied following exposure to a fission reactor environment, as well as the data from the study described in this document. It must be stressed that the completeness of this program and the short time taken to design, build, and implement the program would not have been possible without the dedicated and skillful efforts of scientists, engineers and technical staff at a number of institutions, including Savannah River Technology Center and the Savannah River Site, Lawrence Livermore National Laboratory, Sandia National Laboratories-Albuquerque, Sandia National Laboratories-Livermore, Brookhaven National Laboratory, Oak Ridge National Laboratory, Pacific Northwest National Laboratory, and Los Alamos National Laboratory. Facility development has also enjoyed the expert assistance from staff at Forschungzentrum-Juelich in Germany, the Paul Scherrer Institute in Switzerland, and RISO National Laboratory in Denmark. The authors express their appreciation for the opportunity to represent all these persons in the presentation of this report.

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