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OPERATING EXPERIENCE WITH THE IPNS NEUTRON GENERATING SYSTEM

A. W. Schulke, D. E. Bohringer, B. S. Brown, J. M. Carpenter

Argonne National Laboratory
Argonne, Illinois 60439

Summary

The Intense Pulsed Neutron Source (IPNS) has operated since May, 1981, using two Zircaloy-2 clad uranium targets, their light-water cooling systems and various moderator-reflector systems. IPNS has two independent facilities, the Neutron Scattering Facility (NSF) and the Radiation Effects Facility (REF). This paper describes these IPNS facilities, the Neutron Scattering Moderators, and operational experiences with the systems.

Description of Neutron Generating Systems

The proton acceleration process begins with production of H^- ions in the ion source; these are accelerated to 750 KeV in the Cockroft-Walton preaccelerator and further accelerated to 50 MeV in the linear accelerator. At injection into the rapid cycling synchrotron, two electrons are stripped off in a thin polymer foil and the resulting proton is accelerated to an energy of 400-500 MeV. All of the protons are extracted in a single revolution so that very short (100 nano-seconds) pulses of protons are delivered to the uranium targets. This process is repeated 30 times per second. The proton beam passes through an aluminum vacuum window at the end of the proton transport line into the IPNS neutron generating system. A switching magnet just before the vacuum window diverts the proton beam 5° either to the right or to the left into the chosen uranium target. Between the switching magnet and the targets is a proton beam gate to guarantee exclusion of proton beam from the non-operating target in case of switching magnet failure. Also in front of each target is a proton beam diagnostic well containing instruments (PAS/SWIC) designed to assure that the proton beam characteristics (position and size) are proper before impinging on the targets.

The IPNS Zircaloy-2 clad uranium targets have been in use in both the neutron scattering facility (NSF) and radiation effects facility (REF) since startup (1981). The depleted uranium targets consist of eight 25 mm-thick, 100 mm-diameter uranium alloy disks, clad with 0.5 mm Zircaloy-2, (1.5 mm on circumference) cooled by demineralized water flowing in 1 mm channels between the disks. Disks 1, 3, 5, and 7 contain small, steel-sheathed thermocouples in zircaloy wells at their centers.

The thermocouples in disks 1 and 3 provide diagnostic information. The disks are housed in a 304 stainless steel vessel. The two targets are identical except for a 90° rotation about an axis parallel to the proton beam.

Demineralized water for target cooling is provided by two separate, but identical, closed-loop water systems that transfer target heat to a secondary water cooling system. Each system has a primary pump, heat exchanger, a helium-gas covered surge tank equipped with a hydrogen recombiner, storage tank, ion exchange column, filters, piping and associated valves, as well as necessary control and diagnostic instrumentation. The systems are sealed inside a 6 ft^3 aluminum box. A Geiger counter mounted near the ion exchanger, monitors gross gamma activity primarily due to positron-annihilation and nitrogen 16 gamma rays. In case of target clad failure, the level of accumulated fission products in the target coolant ion exchanger and surge tank cover gas is an indication of the target cladding integrity.

The cover gas from the surge tank is also circulated outside the biological shield to a NaI gamma ray detector and then returned to the surge tank. The circuit provides a 180 minute holdup time which allows decay of the intense short-lived gaseous activity (^{13}N, ^{11}C, etc.) generated by the reaction of protons and neutrons with the cooling water and dissolved gases. The 250 KeV gamma rays associated with the decay of ^{135}Xenon are then measured, whose presence in the gas above the surge tank has been determined to be the most sensitive on-line indication of fission products leaking from the target. The fission yield of ^{135}Xenon is high; the specific activity is high; and xenon, as a gas, moves readily through very small openings in the target clading. The fission product monitor is backed up by grab samples from an on-line gas sampling station. The cover gas in the surge tank exhausts through a holdup device, a HEPA (High Efficiency Particulate Air) filter, the neutron facility ventilation exhaust blower and then exits to the exhaust stack. A stack monitor is located in the exhaust gas stream to monitor the effluent.

The neutron generating facilities and target support systems are surrounded by a biological shield which provides structural support, shielding, and physical protection against damage from external forces. The biological shield consists of an inner steel shield (1100 tons) immediately surrounding the neutron generating system components and an outer concrete shield (50 tons). The steel shield is enclosed in an atmospheric control tank which functions as the boundary of the shield atmosphere control system, which is operated at slight vacuum. The shield is designed to limit radiation dose rate at any accessible location in the experimental area from all radioactive sources within the shield during normal operation to 0.5 mrem/hr. The shielding also keeps the background levels at the neutron scattering instruments to a minimum.

Neutron Scattering Facility (NSF)

In the neutron scattering facility, the fast neutrons produced by fission and spallation reactions in the target are thermalized by moderators (with a high hydrogen density) located above and below the target. We have used two types of moderators. One type consists of four, ambient-temperature polyethylene moderators with an inner graphite reflector. The second consists of four 100 K circulating liquid CH_4 moderators with an inner 100 K beryllium reflector. These moderators are surrounded by an outer, 300 K beryllium reflector. Twelve horizontal neutron beam tubes and one vertical neutron beam tube view selected faces of the moderators to provide neutron beams to instruments. Each horizontal beam tube contains a 36" thick steel beam gate. Two vertical holes (VT3 and VT4) and two horizontal holes (F-6 and H-2) may be used for sample irradiations. A polyethylene insert has been added to one of the vertical holes to enhance the neutron flux and has resulted in an increase in the thermal neutron flux by a factor of two.

Radiation Effects Facility (REF)

In the radiation effects facility, the neutrons emitted from the target pass into high density reflector materials (lead) which maximizes the fast neutron flux within irradiation holes, two vertical (VT1 and VT2) and one horizontal. Thimbles are provided to permit samples to be inserted and removed. Two vertical tubes are equipped for irradiation of samples at controlled temperatures between 4 K and 600 K; the horizontal tube operates only at ambient temperature.

Control and Monitoring Systems

The control system for IPNS consists of 1) a mini-computer for processing information, 2) a hardwired data input/output system for gathering information providing for control and proton beam shutdown and 3) a Cathode Ray Tube display and keyboard. The computer also fills a backup role to the separate hardwired proton beam shutdown system by providing alarms and a software proton beam shutdown.

Moderators

The moderator-reflector assembly used during the first year was of room temperature polyethylene with graphite reflector. This was installed to allow modification of the beryllium-reflector L-CH_4-L-H_2 system, which we discovered would not function as designed.

After 15 months operation, the polyethylene moderators and graphite reflector assembly was removed from the NSF during the August, 1982 shutdown. The poly-ethylene-graphite assembly had three ambient-temperature poly moderators, with inner graphite reflector and outer beryllium reflector, and cadmium decoupling and void liners throughout. This assembly did not provide a vertical beam moderator. Upon removal of this assembly, the radioactivity levels were found to be 40 mr/h to 120 mr/h on the two steel shield plugs above the reflector and 200 mr/h at 5 cm for the graphite-poly assembly. These parts were bagged and stored in shielded containers. The removal procedure was relatively easy with the only surprise being loose rust contamination around the top area of the moderator well. Activity levels ranged up to 2 x 10^3 dis cm^2/min $\beta\gamma$ with no detectable α contamination. These areas were wiped down (wet) and covered with anti-rust paint to reduce the spread of activity. It was thought that the areas where the rust was found were in a helium atmosphere. We investigated the operation and integrity of the helium system. The system was found intact and the helium flow increased. Our best guess for the cause of the rust was that the two concrete and lead shot filled, steel cased shield plugs were not dried long enough to rid the concrete of its moisture and the moderator housing was not sealed properly so as to let air leakage in via the neutron facility ventilation system.

The liquid methane moderators and inner beryllium reflector assembly were checked, tested and cycled three times using its refrigeration equipment before it was installed in the NSF facility. In process of testing, we found that the "L-H_2" systems could not operate at 20 K while the "L-CH_4" systems operate at 100 K. We decided to forgo use of 20 K L-H_2 moderators, and to operate all four moderators with 100 K L-CH_4. All tests then indicated that reliable operation could be expected. The proton beam was put on the NSF target from 29 September to 4 October 1982 (total of 2.17 x 10^{19} proton pulses) with a proton energy of 400 MeV. After two days of beam on target the flow to "H" moderator showed degradation, by the fourth day the flow was blocked. Various attempts to restore flow were tried including cycling the throttle valve (thought to have frozen due to moisture), changing liquid nitrogen flow to the heat exchanger, and bypassing part of the methane flow. All attempts were to no avail. The facility was shutdown and the moderator systems were warmed up. The methane gases were circulated, gas samples were taken and the methane gas was changed. The original methane charge was Grade 4.2: methane (CH_4) 99.992%, nitrogen (N_2) 20 ppm, oxygen (O_2) 4 ppm and ethane (C_2H_6) 12 ppm. The new charge was Grade 4 99.95% methane (CH_4) and would not result in a significant change in impurity levels.

After another attempt at operating the system we decided the problem was the plugging of the 15 micron filters which were located in the weld-sealed, evacuated heat exchanger box. These filters were removed and replaced by 90 micron filters. The new filters were relocated in an accessible part of the system and fitted with easily removed slip couplings. Proton beam was again delivered to the system and flows to the 4 liquid methane moderators at 100 K were much improved. Prior to this run, we also changed the 90 micron filters to assure ourselves that we had cleaned the foreign residue from the system (see picture). Samples of the methane gas were also taken again.

This photo is a magnification (70X) focusing on large particles in the upper right quadrant. Light colored regions indicate a non-conductive material.

From 7 December to 21 December the system ran with proton beam on target; the 4 methane moderator systems ran with no problems. A significant increase in the neutron flux was seen (factor of about 8 at 5Å). Experiments that were run at this time indicated that the larger moderator, (thicker in the direction of the neutron beam) resulted in an increase in time width of the pulse that degraded the resolution in the powder diffractometers. This led to the decision that the moderators would be reworked. During the last operating cycle on the methane system, we erected a vacuum line to test the solubility of various boron compounds in cryogenic moderator fluids to seek an easy fix to the degraded resolution problem on the powder instruments. No practical solution was found. Methane gas samples were taken after the run.

Table 1 - Summary of Mass Spectrometer Measurements on Irradiated CH_4

Irradiation Conditions	Moderator	Mole % CH_4	Mole % H_2	Mole % C_2H_6	Mole % C_3H_8	Mole % C_4H_{10}	Mole % Other Hydrocarbons (C_n,n>4)	Mole % H_2O
After 2.2 x 10^{19} protons on target	H	99.6	-	0.3	0.1	-	<.02	<.05
	F	98.8	-	0.9	0.2	-	.08	.02
	C	99.6	-	0.3	0.1	-	<.02	<.03
	V	99.3	-	0.5	0.2	-	<.02	$\leq$.03
After 5.2 x 10^{18} protons on target	H	99.2	<.06	0.4	0.2	0.1	-	<.03
	F	99.7	<.06	0.2	0.08	0.02	-	<.03
	C	99.1	<.06	0.5	0.3	0.1	-	<.03
	V	99.3	<.06	0.5	0.1	0.02	-	<.03
After add'l 4.6 x 10^{19} protons	F	99.8	-	0.2	0.03	0.003	-	<.03
	V	99.8	-	0.2	0.01	0.002	-	<.03
After add'l 4.8 x 10^{19} protons	H	98.0	1.5	0.4	0.06	-	$\leq$0.02	-
	F	97.9	1.5	0.5	0.06	-	0.02	-
	C	99.1	0.7	0.2	$\leq$0.02	-	<0.03	-
	V	98.4	1.2	0.3	0.04	-	0.04	-

Prior to changing of the in-line filters, there was a concern that flow problems were due to possible heavy hydrocarbon buildup, but the gas samples taken at various times proved that this buildup was minimal. The mass spectrometer analysis of the methane prior to irradiation was CH_4 99.9, $H_2O \leq .02$, $N_2 + CO < .01$, O_2 .005, Ar < .005, $CO_2 < .005$, H_2 0.0, C_2H_6 0.0, C_3H_8 0.0, C_4H_{10} 0.0, other hydrocarbons $\leq$ .05. These numbers are in mole percent. The results of the analysis after irradiation are given in Table 1. ANL's chemical analytical lab ran tests on the used filters and recommended a cleaning agent for cleaning hydrocarbon buildup if needed. The most effective solvent for cleaning seems to be tetrahydro-furan.

One other minor problem was the activity level of ^{11}Carbon that built up in the heat exchangers during operation. Activity levels up to 250 mr/hr $\beta\gamma$ were measured. If this system would operate continuously, shielding would need to be added around the heat exchangers.

The polyethylene graphite beryllium moderator assembly was modified to provide a vertical moderator (LN_2 cooled polyethylene) for the ultracold neutron experiment. Also, new polyethylene moderators replaced the ones inserted August 1981. This system has been in operation since February 1983 till August 1983.

During this summer shutdown, August-September 1983, we are building a new graphite/polyethylene reflector moderator system. The new system will have an $L\text{-}N_2$-cooled polyethylene moderator in the "C" position. This should provide a higher intensity of long wave neutrons to the SAD, ^{3}He and polarized neutron instruments.

The removal of the methane/beryllium moderator from the NSF went well; levels of radiation were 2.1 R/h at 5 cm $\beta\gamma$. A portable hyperpure germanium detector and 8 K multichannel analyzer were used to identify predominent radioactive nuclides. Using the observed relative intensities and known values for exposure rate per unit activity and gamma's per desintegrations, the relative contribution of the observed nuclides was calculated. Using the respective values for half-life, the time dependence of the radiation field was estimated for several points in time up to one year. The exposure rate is expected to decrease by $\sim$ 18% the first month and $\sim$ 6 % for the first year. Monthly readings taken by the radiation safety people on the moderator has shown these calculations to be fairly accurate. The moderator as of August 1983 reads $\sim$ 565 mr/h $\beta\gamma$. At this time we have not decided whether or not to rework the moderator seen by the powder diffractometers.

Primary Water Cleanup Ion Exchange System and Water and Gas Sampling Program

Each of the two target cooling systems has a prefilter made up of three (3 micron) cartridges. Approximately 25% of the total system flow (50 gpm) goes through this filter housing with the remaining flow through a bypass line around the filter.

Down stream from the pre-filter is a motorized valve which is set to divert some of the water (1-10 gpm) thru a mixed resin bed. The resin column contains $\sim$ 1.9 cubic feet of Illinois Water Treatment #NR-6 (HOH) resin. Part of the total flow is also diverted to a water quality-sample station with an on-line conductivity instrument then returned to the main flow stream. Water samples for checking Ph and radioactivity can also be taken at this station. All primary flow then passes through an after-filter made up of three (3 micron) sintered stainless steel cartridges. This after-filter's primary function is to stop any resin that might escape from the ion exchange which could possibly plug the target flow channels. The resin in the ion exchangers has been in the system since startup of IPNS. These resins have kept the deionized water used in the primary system at a conductivity level below .1 μmho's and a Ph of $\sim$ 6.8.

Since we are concerned with the U target and the possibility of a target clad failure, we decided to periodically remove a sample of resin from the ion exchanges so it could be analyzed for uranium content. The methods used are (EDXRF) x-ray fluorescence and gamma-ray analyses. The fluorescence method is the most sensitive. A sample of factory-fresh resin showed 20 nanograms/gram uranium by x-ray fluorescence and none detected by gamma-ray analysis. Samples of

the system resins ranged from 22 to 53 nanograms/gram over a two year period. Gamma-ray analyses showed various isotopes that one might expect ^{7}Be, ^{46}Sc, ^{51}Mn, ^{57}Co, ^{58}Cu, and ^{88}Y, but no detectable fission products. We feel this is a good operational diagnostic tool.

The primary water samples that are taken periodically are analyzed for radioactive nuclides dissolved or suspended in the water. After several hours decay, the most significant activity in the water is ^{7}Be, and ^{3}H. Other nuclides in much lesser amounts are detected but the purpose of these analyses are for evidence of target clad failure. These analyses have also given no evidence of target clad failure. Lead shielding was added around the ion exchangers and dense concrete shielding put on top of the primary water coolant boxes to reduce the high gamma-levels produced primarily from ^{7}Be from the water (i.e., spallation product).

The gas sampling program has also been conducted since startup of IPNS. Gas samples from the primary coolant cover gas are taken periodically during operation. The samples are analyzed for radioactive gases and act as a backup to the automatic fission product monitor. These samples are also analyzed for any possible hydrogen buildup in the cover gas. The analysis of the grab samples has shown presence of ^{76}Kr, ^{77}Kr, ^{79}Kr and ^{41}Kr. The samples are analyzed after the very short half-life nuclides such as ^{13}N, ^{16}N, ^{15}O, and ^{11}C are allowed to decay. No other gaseous nuclides have been detected. Typical concentrations of ^{77}Kr are $5 \times 10^{-5}\mu Ci/ml$ and ^{41}Ar $1 \times 10^{-4}\mu Ci/ml$.

A mass spectrometer determines H_2 concentration. The H_2 concentration is typically less than 0.5%. The experience with hydrogen buildup that we encountered with the Zing-P' cooling system has not occurred with the IPNS cooling systems. Possible causes for elimination of the hydrogen buildup are the HOH resins that we now use, the better gaseous mixing device installed in the surge tank, the addition of pre-heating the palladium-coated aluminum oxide pellets in the recombiner, and the Zircaloy-2 clad of the uranium. Therefore it remains a mystery why we found the excess hydrogen at Zing-P' and essentially none at IPNS.

Uranium Targets

The uranium targets have functioned trouble free. Two situations have occurred that are worth mentioning. During the early testing of the cold methane moderators the coolant flow to the NSF target was shutdown for the night. A casual check of target temperatures indicated that they were very close to freezing. This happened because a thermal short developed between the 100 K surfaces in the moderator-reflector assembly and the outer surface on which the target housing rests. The moderator systems were shutdown for the night and allowed to warm up. We then visually inspected the target to assure it was not damaged and took photographs of the target. Target flow was reestablished and various target system parameters were checked. One should make sure that standard operating procedures are established when the target coolant systems are operating and the cold moderators are operating. An option is to install an interlock between the two systems.

The second operating experience that occurred recently (within the last few months) was that two thermocouple readouts to each of the targets opened. We investigated this during our present shutdown and discovered corrosion at the thermocouple connection. These connectors were cleaned and the readouts returned to normal. We hope to prevent reoccurrences of this problem by adding an interlocking neoprene boot around the connector, and also bagging the connector in an inert atmosphere. The junctions themselves, within the target disks, were not involved nor affected.

Proton Beam Diagnostic Systems

The purpose of this system is to provide safe beamline operation and to prevent "hot spots" on the targets. The beamline performance is assessed by a position and size (PAS) monitor (ionization monitor) and a segmented wire ionization chamber (SWIC). The PAS produces proton beam shutdown signals for beam too small, too large or off axis. The SWIC produces a separate signal to obtain two profiles from which are obtained the beam size and position in the horizontal and vertical planes. Each target is equipped with a PAS and SWIC in the diagnostic well.

These monitors have been very reliable but as mentioned in previous sections of this paper, corrosion in these wells had been a problem. These wells are made of steel and have a helium supply system. The helium supply was inadequate, so an additional helium line was added to directly blow inert gas on the diagnostic monitors. The steel shield plug was sprayed with molykote molybdenum disulfide compound to decrease rust and other corrosion buildup.

The point to be made here is that during construction stages the steel should be coated with some anti-corrosion agent, painted or even electro-plated. This would make life much easier for operations personnel who have to maintain equipment located in these environments.

Stack Monitoring System

The IPNS stack monitor system serves three purposes. Primarily, it provides real time information on the radioactivity being released up the stack along with alarm capability to alert operating personnel to a problem. Second, it provides record documentation to support required reports of the total quantity of radio-nuclides released to the environment in the gaseous effluent from the IPNS. Third, this system provides a measure of backup monitoring and alarm capability for the target coolant and the ion exchange resin monitoring systems to trigger an alarm in the event of a major release of radioactive nuclides through these systems.

The IPNS stack monitor is composed of a set of radiation detectors on a sampling line designed to quantitatively measure the radioactive gases and particulates in the stack effluent of the IPNS. The system includes the air pumps, flow control, signal processors, data readouts, power supplies and piping, and electrical interconnection between components. The air pump pulls a continuous sample from the stack effluent stream, downstream of the HEPA filters, and passes it sequentially through a particulate filter, an activated charcoal gas adsorbent, and a noble gas counting chamber before returning the sample flow to the stack. The sample flow rate is controlled to be isokenetic with the flow of the effluent stream. The materials collected on the particulate filter are counted for β activity with a plastic β scintillator (50 mm diameter x 4 mm thick)/photo tube assembly. The activated charcoal adsorber is continuously counted for γ activity with a NaI(Tl) (50 mm x 50 mm) scintillation detector. The $\sim$300 cm^3 noble gas chamber is continuously counted for β activity with a similar plastic β scintillator. All three detector channels are provided with background compensation to

reduce the effects due to changes in the ambient γ ray background. Each detector is shielded by a minimum of 5 cm of lead in the form of fine shot contained in a welded steel box.

The output data from each counting channel is sent to the IPNS central control CAMAC system, at one minute intervals. The display readout is provided in the IPNS main control room either on operator command or whenever one or more alarm points are exceeded.

The readout is printed at the terminal once each hour when all operating conditions are within normal range. Under abnormal conditions, e.g., an alarm point exceeded, the channel data is printed once each minute. The format for the channel data printout is digital and graphic in form with time of day and six channels of data being printed and the data graphed on the same line of print

The stack monitor is a good diagnostic tool for operations personnel. After background and running limits are set, operations personnel can also be alerted to changes various experimenters make on the instruments, i.e., off gassing from pumps, warming up and pumping out of cryostats etc. The stack monitor can also act as a backup to the fission product monitor.

Conclusions

The Neutron Generating Systems have operated without serious trouble since the startup of IPNS. The original cold moderator system as designed would need extensive changes, especially to the liquid hydrogen moderator hardware. The available sub-cooling was also marginal for the liquid methane moderators. Therefore, with the prospect of an increased heating load from an enriched uranium target, a new cold moderator system will be designed and built.