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- Subject: Project No. 704 BWRVIP-96NP: BWR Vessel and Internals Project, Sampling and Analysis Guidelines for Determining the Helium Content of Reactor Internals
- Reference: Letter from Carl Terry (BWRVIP) to Document Control Desk (NRC), "Project 704 BWRVIP-96: BWR Vessel and Internals Project, Sampling and Analysis
 Guidelines for Determining the Helium Content of Reactor Internals," dated November 29, 2001.

Enclosed are two (2) copies of the report "BWRVIP-96NP: BWR Vessel and Internals Project, Sampling and Analysis Guidelines for Determining the Helium Content of Reactor Internals," EPRI Technical Report 1003019NP, May 2002. This is a non-proprietary version of the proprietary document submitted to the NRC by the letter referenced above.

If you have any questions on this subject please call Robin Dyle (Southern Nuclear, BWRVIP Integration Committee Technical Chairman) at 205.992.5882.

Sincerely,

William A Eatur

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BWRVIP-96NP: BWR Vessel and Internals Project Sampling and Analysis Guidelines for Determining the Helium Content of Reactor Internals

Technical Report

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BWRVIP-96NP: BWR Vessel and Internals Project

Sampling and Analysis Guidelines for Determining the Helium Content of Reactor Internals

1003019NP

Final Report, May 2002

EPRI Project Manager R. Thomas

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REPORT SUMMARY

The Boiling Water Reactor Vessel and Internals Project (BWRVIP), formed in June 1994, is an association of utilities focused primarily on BWR vessel and internals issues. This BWRVIP report provides guidelines for removing a small sample from a reactor to measure helium content (for weldability determinations) as well as other parameters.

Background

One of the factors affecting the weldability of stainless steel is the presence of helium in the material. Helium is produced by the transmutation of boron and nickel when they interact with thermal neutrons. If the helium content is low enough, welding can be performed by conventional techniques. However, if helium content is high, welding requires the use of special techniques, or in some cases, cannot be performed. Utilities require a method for determining helium content of in-situ materials. One method involves removing a small sample of the subject material from the reactor and subsequent spectrographic analysis.

Objective

To develop guidelines that describe acceptable methods for sample removal and analysis of helium content.

Approach

As part of a separate effort, a joint BWRVIP/NRC project was conducted to remove samples from three BWRs and to analyze the samples for helium content as well as for other parameters. Lessons learned from this effort allowed the project team to develop guidelines for effective removal and analysis of an appropriately chosen sample.

Results

The resulting guidelines include instructions for removing the sample, transmitting the radioactive material to the analysis laboratory, specifying the required analysis, and performing a safety evaluation to demonstrate that the sample removal does not create a site that will promote crack initiation. In addition, the guidelines present sufficient details of the sampling that was performed as part of the BWRVIP/NRC project to give utilities an appreciation for the scope of required activities.

EPRI Perspective

The report presents guidelines that will facilitate utility efforts to measure helium content in a reactor material for purposes of determining weldability. In addition to determining weldability, additional, straightforward sample analyses can be performed to determine chemical composition as well as fast and thermal neutron fluence. These latter measurements are useful in benchmarking fluence calculations at the vessel or at other internal locations.

Keywords

Boiling water reactor Vessel and internals Repair Fluence Helium Welding

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1 INTRODUCTION

One of the factors that affect the weldability of stainless steels in BWR reactors is the helium content of the metal. Helium is produced when thermal neutrons interact with boron and nickel. The resulting nuclear reactions leave behind a quantity of helium that becomes trapped in the material. When the material is melted during welding, the helium is released and forms small bubbles. If the concentration of helium is above a certain threshold (typically about 0.1 appm), the bubbles will be produced in sufficient quantity to degrade the strength of the material and to cause cracks in the weld heat affected zone. Consequently, if welding is to be considered as a means of repairing an irradiated reactor component, it is important to know the approximate helium content at the weld location.

For some internal reactor components, the helium level can be calculated. This involves estimating the fluence at the location as well as knowledge of the boron and nickel concentrations in the material. While this process is straightforward, the calculated helium concentration may be subject to substantial uncertainty. There is uncertainty in the thermal fluence calculations due to the fact that they have not been accurately benchmarked at many locations of interest (e.g, jet pump riser brace pads or core spray piping). In addition, the boron concentration of the subject material may not be known. Boron was considered a "tramp" element and was not typically recorded as part of the material certification process. Its concentration may vary from less than 1 ppm to over 50 ppm. Consequently, a more direct and accurate method for determining the helium concentration is needed in some cases.

One acceptable method for determining the helium concentration is by direct measurement on a small sample removed from the component of interest. The amount of material required for the analysis is small (<50mg) and the determination of helium content by mass spectroscopy is straightforward and yields accuracies on the order of 1-percent.

Recognizing that utilities may need to perform such sampling and analysis in the future, the BWR Vessel and Internals Project (BWRVIP) and the US Nuclear Regulatory Commission (NRC) conducted a joint project to demonstrate sample removal from a number of BWRs. Samples were removed from the jet pump riser brace pad at four locations in three US BWRs using tooling developed by Framatome – Advanced Nuclear Power (FRA-ANP). The samples were sent to Pacific Northwest National Laboratory (PNNL) where they were analyzed for helium content, initial boron content and accumulated fluence. The results of this project are further described in Reference 1.

The objective of this Sampling and Analysis Guideline is to use the lessons learned from the BWRVIP/NRC project to provide utilities with guidance on performing similar sampling on internal components. Section 2 of the report discusses the basic requirements for removing the sample and obtaining the desired analyses. Section 3 presents an overview of the

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BWRVIP/NRC jet pump riser brace sampling. It is intended to provide utilities with a better understanding of the scope of the process, a description of typical tooling, sample handling requirements, requisite safety analyses, etc.

The discussion in this report is based primarily on experience using the Framatome tooling and analyses performed by PNNL. However, other methods of sample removal and analysis are also acceptable for determining the helium content of stainless steels for purposes of assessing weldability.

2 SAMPLING / ANALYSIS REQUIREMENTS

This section presents the basic requirements for removing and analyzing a sample including required sample size, sample handling, specification of the required analyses and required safety evaluations.

2.1 Sample Requirements

2.1.1 Sample Size

In order for the laboratory to perform a complete analysis including determination of helium, boron and fluence, a sample of sufficient size must be provided. The sample mass must be at least 20 milligrams. 50 milligrams is preferable. The samples can be in the form of chips, shavings, chunks or strips. The only limitation in form is that the individual chips in each sample need to be at least 1-2 mg. to perform the required analysis. The picture below shows the sample collection filter containing the material taken during a test of the sample tooling. The filter is approximately 1" in diameter. This shows the representative size of actual samples for one location. This sample was produced from a 1/4" diameter drill bit at a maximum depth of 0.06".

The sample should be removed from a location in close proximity to the potential weld repair location and should consist of the same material on which the weld will be placed. If possible two samples should be removed in order to average out spatial variations in helium content. In selecting locations for sampling, consideration should be give to the shielding effects of nearby obstructions that could cause the fluence (and consequently the helium content) of the sample to differ from that of the material at the weld location.

Sampling / Analysis Requirements



Figure 2-1

2.1.2 Sampling Process

The jet pump riser brace pad samples from the BWRVIP/NRC project were obtained by drilling a small divot (1/4-inch diameter by approximately 0.06-inch deep) in the material of interest and collecting the drill shavings. The tooling used for this process is described in Section 3. The tooling will likely have to be modified (or alternate tooling developed) in order to allow sampling at other plants. Drawings and videos are required for the tooling and mockup designs. After fabrication, the tooling must be tested and qualified. A site procedure to take the samples must be prepared and the tooling should be tested on a full scale mockup using the site procedure.

Using the Framatome tooling, the sample removal process can be performed remotely from either the refuel or auxiliary bridge with the use of long handled poles. Since the work is performed off a bridge, the exposure levels are low. The work over the vessel is typically performed in two 12-hour non-continuous shifts. The actual schedule will depend on what other activities are being performed at that time. Foreign material exclusion (FME) must be employed during all aspects of the sample removal process. The area to be sampled should first be video

inspected to determine if the area requires cleaning to remove loose contamination, and to spot any obstructions that may hinder the sample removal process. Brushing or hydrolazing can be used for this cleaning. The sampling area should be video inspected after the sampling to determine if the sample area is the same configuration as that which was qualified. The details of the tooling, qualification, and the process are described in Section 3.1.

The equipment for sample removal included the following:

- Sample removal tooling
- Underwater video camera suitable for installation and pre/post inspection
- Hydrolazing wands and hoses (Plants supplied the hydrolazing pump), if the sampling location requires to be cleaned
- Support equipment (handling poles, VCR, monitors, videotapes, etc.)
- Sample shipping container(s)

2.1.3 Sample Handling / Shipping

Typically samples of this type are shipped in a lead lined steel container (AKA "pig"). The size of the pig is determined by the dose rate of the samples. The requirements for shipping LSA material are contained in 10CFR49. If the dose rates are low (as was the case in the BWRVIP/NRC project), the samples can be shipped as a limited quantity shipment through an express mail service like Fed-Ex. The requirement (10CFR49 Part 173) for shipping limited quantity is that the outside of the container is reading less than 0.5 mR. If the samples have a higher dose rate, a "pig" will have to be used. The shipper must also verify that the samples are within the limits of the analysis laboratory's license.

2.1.4 Definition of Desired Analysis

The samples may be analyzed for helium content, boron content, fluence (fast and/or thermal) and general chemical composition. Helium content is the primary measurement required for determining weldability. Boron may be useful if, in the future, it is contemplated that analytical methods may be used for determining helium content of similar heats of materials. Measured fluence is useful as a benchmark for future analyses and also provides direct information that can be used to "calibrate" reactor physics codes.

The following language may be used to specify the helium, boron and fluence analyses to the laboratory:

Measure the helium and boron contents in each sample using mass spectrometric methods of small specimens taken from the drill fillings provided. The specimens shall be chosen, either using optical methods, or by gamma counting, to be representative of the bulk of the material. Specimens with surface irregularities, or with evidence of oxidation, should not be used for the analyses. Helium determinations will be conducted by gas mass spectrometry. Results will be reported as atomic concentrations (appb ⁴He) relative to the total number of atoms in the sample to an accuracy of approximately 2%. Boron

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measurements will be conducted by determining the increase in helium content from the ¹⁰B (n, α) ⁷Li reaction after supplemental irradiation of a sub-set of the samples in a thermalized neutron environment. Determination of the thermal neutron fluence will be made via the ⁶Li (n, α) ³H reaction using well-characterized Al-⁶Li alloy included in the irradiation assembly, or by using other accepted thermal neutron dosimeter materials. Natural Boron (NB) measurements will be reported as weight concentrations (wt.ppm NB) to an accuracy of approximately 5%.

Characterize the neutron spectra at the location of each sample using retrospective dosimetry methods. The measurements will include compositional analysis by X-ray fluorescence, and radiometric counting to determine the activities of ⁵¹Cr, ⁵⁶Co, ⁶⁰Co, ⁵⁴Mn, ⁵⁹Fe, and ⁶³Ni, ⁵⁵Fe, and ^{93m}Nb, as appropriate. Cobalt impurity concentrations in the samples will be determined from gamma analysis of the samples irradiated as part of the boron determinations. The radiometric and composition data will be used to determine the fast and thermal neutron fluence exposures. If the compositional analysis indicates sufficient ⁹³Nb, the activity of ⁹³mNb will also be reported and used in the fluence determinations. Thermal and fast (> 0.1 MeV and > 1. MeV) neutron fluences will be reported as neutrons/cm2 to an accuracy of approximately 10% or better.

Assuming that prior contractual arrangements have been made, results of the helium analysis and an estimate of the initial (i.e., pre-irradiation) boron content of the samples can be available within approximately one week of receipt of samples by the lab. Complete analysis for fluence and chemical composition may require as much as one to two months.

2.2 Required Safety Analysis

A technical justification is required to support development of a safety evaluation for the sampling activity. The technical justification should address testing, qualification and use of the equipment as well as relevant structural and material issues associated with the as-left condition of the sampling location(s).

The removal of the sample will result in a stress concentration that was not considered in the original design calculations. The following discussion shows how the effect of this stress concentration on the fatigue life of the sampling location can be addressed so that the rules of ASME Section III are not violated.

The existing stress report and thermal cycle drawing must be reviewed. Not all the stresses required to perform the fatigue analysis may be contained in the stress report. Supplementary analysis may be required to calculate other required stresses for the fatigue analysis. The significant transient loads considered in the original calculations are:

- Design Hydrotest
- Sudden Heatup and Sudden Cooldown
- Normal Heatup and Normal Cooldown

The new peak stresses and fatigue life must be calculated at the location of the sample removal for the transients that contribute to the usage factor. This is accomplished in the following steps:

- Calculate stress concentration factor (SCF) caused by sample removal
- Apply SCF to range of primary + secondary + peak stresses
- Calculate revised usage factor

A detailed discussion of the structural analyses performed for one plant in the BWRVIP/NRC program is contained in Section 3.2.1.

A material evaluation of the divot in the sampling location must be performed. The cold worked surface layer due to machining must be evaluated. The crevice condition that will exist in the machined divot must also be evaluated. The detailed description of the material evaluation is contained in Section 3.2.2.

The plant should consider if there is a need for an independent reviewer for the safety analysis.

2.3 Plant Information Required

The following plant specific information is typically required for the design of the tooling and mock-ups, and the engineering technical justification for the sampling process. Additional information may be required:

- Stress report (including stress/load changes due to power uprate)
- Reactor thermal cycle drawing
- General RPV and internals arrangement drawings
- Detail drawings of the location(s) to be sampled
- Configuration and materials of construction of the location(s) to be sampled
- Video footage (IVVI and general area footage) of the location(s) to be sampled
- Drawings which may identify potential obstructions
- Drawings of other components in the region of the sampling location(s)

2.4 Overview of Helium Analysis Procedure

The following is a summary description of the technique used by PNNL for helium analysis.

2.4.1 Definitions

Materials received for helium analysis are prepared as described below (or as specially requested), with portions of the sample being cut into " specimens" for analysis. Except as noted, the sampling includes two independent helium analyses of a pair of specimens cut by the sampling lab from adjacent locations in a single solid sample. This sampling includes cutting, cleaning, and also etching the pieces, if requested, to remove material that could have enhanced or depleted helium due to recoil (the distance an α particle travels from when it is generated until

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it comes to rest) or other effects. Helium recoil can be a significant surface effect for samples which were irradiated in a helium environment (i.e., helium knock-on), or for samples where the produced helium has high recoil energy (i.e., accelerator environments). Maximum recoil distances for fusion reactor environments are typically a few tenths of a mil (i.e., 0.0001 to 0.0002 in.). For the type of sampling discussed in the present report, where the material has been collected from the bulk material beneath the component surface (e.g., by drilling), then the effects of recoil and surface contamination are not a factor, and therefore no etching is required.

2.4.2 Sample Preparation

Upon arrival at the sampling lab, the sample packages are identified and compared with the customer's correspondence. Each sample is then examined to check for adhesions or other surface irregularities, which might affect the helium analysis results. As discussed above, specimens should be selected to be representative of the bulk of the material. Following examination, two smaller specimens are then taken from each sample for duplicate helium analysis. Each specimen is ultrasonically cleaned in alcohol and air dried. The specimens are weighed with a 1 σ uncertainty of less than 1% of the mass value (generally to \pm 0.002 mg), using a mass balance with calibration traceable to the U. S. National Institute of Standards and Technology (NIST).

2.4.3 Helium Analysis Procedure

Determination of the helium content in a sample is performed by vaporizing the sample in a resistance-heated graphite or tungsten-wire crucible in a vacuum furnace connected to a mass spectrometer. The absolute amount of ⁴He released is then measured with respect to a known quantity of added ³He " spike. " Each helium spike is obtained by expanding and partitioning a known quantity of gas through a succession of calibrated volumes. The mass spectrometer is calibrated for mass sensitivity during each series of runs by analyzing known mixtures of ³He and ⁴He.

2.4.4 Accuracy

The amount of helium that can be measured with an absolute accuracy of ~ 1 % ranges from ~ 10^{11} to 10^{17} atoms. Lower or higher helium levels can be measured with some potential reduction in accuracy, but the lower limit is usually set by the background helium released by the vaporization process. The uncertainty in this background ranges from ~5 x 10⁸ to ~5 x 10⁹ atoms of ⁴He, depending on which furnace type is used to vaporize the sample.

For comparison, a 1 mg sample of steel with 0.1 appm helium contains approximately 10^{12} atoms of helium.

2.4.5 Retaining Unused Sample Material

Usually, additional sample material remains after the specimens are taken. This material is normally held for a period of 6 months in case additional analyses are requested. This also depends on the lab performing the analysis.

3 BWRVIP/NRC JET PUMP RISER BRACE PAD SAMPLING

As discussed previously, the BWRVIP and NRC have removed samples from the jet pump riser brace pad (Figures 3-1 and 3-2) at three reactors and have had the samples analyzed for helium, initial boron content, and fast and thermal fluence. An overview of that sampling and analysis is presented in this section to provide utilities with an appreciation for the scope of the process, a description of typical tooling, sample handling requirements, requisite safety analyses, personnel and time requirements, etc.

3.1 Sampling Hardware Description

FRA-ANP developed the sample removal tooling and provided the necessary personnel and equipment to support the removal of samples.

3.1.1 Sampling tooling

The tooling was designed for removal of a sample from the pad on the reactor vessel to which the jet pump riser brace is welded. It was desired that the tooling be useable at a number of different plants with minor modification. During the tooling development, a number of BWR plants provided drawings and footage from in-vessel visual inspection (IVVI) for the riser brace and pad configurations.

The equipment for sample removal includes the following:

- Sample removal tooling
- Underwater video camera suitable for installation and pre/post inspection
- Hydrolazing wands and hoses (Plants supplied the hydrolazing pump)
- Support equipment (handling poles, VCR, monitors, videotapes, etc.)
- Sample shipping container(s)

The sampling tool (see Figure 3-3) is a remotely operated drill fixture that can be lowered into position using handling poles from the refuel or auxiliary bridge. The fixture is secured to the riser brace leaf via a rotating hydraulic clamp arrangement. The pneumatic drill motor carries the sample removal bit. A hydraulic feed cylinder is used to feed the drill motor to and from the pad. Plant de-mineralized water was used in all hydraulic cylinders so as not to affect plant water chemistry in the unlikely event of a leak.

A sheath protects the vessel internals (and bit) during tool installation. It also acts as a hard stop to control the depth of cut and provides containment of the sample. Sharp tool bits are used for each sample removal to minimize work hardening of the material. During the sampling process, a vacuum generator (a simple water powered venturi) is used to create a suction in the annular region between the bit and the sheath. The small chips created are pulled through the sheath into a filter cartridge so as not to create a foreign material exclusion (FME) concern. The following methods are used to determine that the vacuum generator is operating properly. The flow indicator is monitored before and during the cut to verify that proper suction is maintained. The operators ensure that the wheel turns at a comparable speed to that identified during qualification testing. If not, the cut is not started (or the cut will be stopped if previously started). There is also an air bubbler near the inlet of the debris sheath. The bubbler provides a redundant method for verifying proper suction. By visually verifying that the bubbles enter the sheath, it demonstrates that the debris is being sucked into the debris sheath during the cutting operation. Also to determine that the vacuum generator is receiving the proper flow, a pressure gage is installed on the water supply line to the tool. This pressure is monitored just before and during the cut to ensure that the pressure is greater than that used during qualification.

3.1.2 Testing and Qualification

Bench testing of the sample removal tooling was performed on mockups with representative jet pump riser brace (JPRB) configurations. In addition to bench testing, the test plan included demonstrating that the tooling can be remotely delivered using a full height mockup (Figure 3-4). Also demonstrated during testing was the ability to adequately contain the sample in the filter cartridge, eliminating the samples as an FME concern.

The sampling process was performed in accordance with a qualified procedure. Weld pad mockups were manufactured using a weld buildup on low alloy material. Both Alloy 182 (ENiCrFe-3) and Type 308L/309L weld pads were constructed with representative surface finishes. The weld pads were deposited in the flat position using qualified welding procedures. The Alloy 182 material was deposited using manual shielded metal arc welding technique. The 308L/309L material was deposited using a manual gas tungsten arc welding technique.

The weld pad mockup materials used for the qualification were in an un-irradiated condition.

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The qualification plan required that the sampling process leave no crevices (see Section 3.2.2) on the weld pad mockups. After sampling, both PT (color contrast-solvent removable method) and visual inspections (EVT-1) were performed on the divots in the weld pad mockups. The acceptance criteria allow no indications of a crevice being generated as a result of the sampling process. Photographs and video of the qualification samples were taken so that they may be compared to the as-left condition of the riser brace pads on site.

3.1.3 Sample Removal Process

The sampling process involved removal of a small amount of material from the surface of the JPRB weld pad and resulted in a small "divot" being left. The configuration of the "divot" is shown in Figure 3-5. The sampling process was performed in accordance with a qualified procedure. Prior to sample removal, the pad may be either brushed or hydrolazed (cleaned with high-pressure water) at the option of the utility and was visually inspected (EVT-1) in the region where the sampling is performed.

A new filter was placed in the tool. The sampling tool was lowered into position using handling poles from either the refuel or auxiliary bridge. The fixture was secured to the riser brace leaf via a rotating hydraulic clamp arrangement.

Once the sample has been taken, the fixture was removed from the vessel and the filter cartridge was detached, placed in a suitable shipping container and shipped to a lab for processing. Approximately 100 to 200 mg of material was removed from each location sampled. A final visual inspection (EVT-1) of the sampled region was performed to ensure that the as-left condition is comparable to the qualification samples.

3.1.4 Sample Site Support

The utility supplied the following general site support:

- Office facilities for 4 people with phone, fax, modem and photocopier available
- Mobilization of equipment to and from the refuel floor
- Health Physics
- QA support (as required by site)
- Qualified bridge operators (as required by site)

The utility supplied the following Site supports for the refuel floor:

- Equipment laydown/setup area (Approx 15' x 15')
- Clean/dry plant air (10 scfm @ 80-100 psi) (Air which has been filtered and passed through a moisture separator.)
- Demin water supply 40 psi (8-10 gal/min)
- Demin water for hydraulics (2-5 gallons)
- 110V, 20A, 60 Hz power (on bridge and in setup area)
- Site supplied consumables (duct tape, Loctite, etc.)
- Sample shipment

3.1.5 Sampling Personnel

A single four-person crew consisting of one Task Lead, two Reactor Services Technicians and one Tooling Engineer were supplied to support the sampling activities. With this crew, the sampling activities can be performed over a two-day period working two 12-hour noncontinuous shifts. In addition, one 12-hour shift was required before and after the sampling effort for equipment setup and demobilization. If the Plant has two work platforms available, it is possible that the sampling activities could be performed in parallel with other in-vessel activities.

3.1.6 Sampling Locations

Six JPRB pad locations were selected for each of the three plants in the BWRVIP/NRC project. Three locations had high fluence levels and three locations had low fluence levels. Two locations were for contingency in case a sample could not be obtained from the primary locations.

3.1.7 Sample Shipment

The samples taken from each of the three plants had dose rates of <2mr. Since the dose rates were so low, it was possible to ship the samples to PNNL as a limited quantity shipment by Fed-Ex.

3.1.8 Lessons Learned

In performing the sampling at the three plants, there were several lessons learned that were incorporated into the project. Some factors that were incorporated into tooling modifications include: a secondary means was needed to ensure that the vacuum was operating properly; a flow meter was added to the water supply side of the vacuum and a bubbler was added in front of the sampling tube to ensure that the sample chips would be sucked into the sampling tube.

After the first plant was sampled, the need for hydrolazing was revisited. There was very little difference in the surface condition before and after hydrolazing. Therefore, it was not performed at the two other plants. There was no difference in the dose levels of the samples with and without hydrolazing.

Foreign material exclusion (FME) is always a concern when operating tooling inside the reactor vessel. Tooling needs to be designed to ensure that parts do not become a FME concern. An example of this is that a quartz window of a camera light had become separated from the housing causing the reflector light assembly inside the housing to also become separated and lost in the Reactor Vessel. The thoughts are that the epoxy, which seals the lens to the housing, had degraded after time and had failed. The housings are now being visual inspected for signs of degradation. Tape is also being placed over the outer edges of the lens to secure the lens to the housing to prevent a lost parts situation even if the lens were to fail.

A concern was raised regarding cross contamination of the samples. To a certain degree, the location of the sampled chips can be determined from the measured fluence level. However, in order to avoid resorting to this means of sample identification, the tooling should be checked to ensure that chips are not left in the tool from the previous sampling. As a minimum, a visual inspection should be performed; a more positive verification can be performed by flushing the lines.

3.2 Safety Analysis

A safety analysis was conducted to ensure that the sampling would not leave the surface in a degraded condition. The safety analysis consisted of a structural analysis and a material evaluation of the sample divot in the JPRB weld pad. The safety analysis was reviewed by an independent reviewer, Structural Integrity Associates (SIA). While the analyses performed for the three plants differed somewhat, details of one safety analysis are presented below to indicate the types of evaluations performed.

3.2.1 Structural Analysis

Removal of the sample from the weld pad will result in a stress concentration that was not considered in the original design calculations. The following discussion demonstrates that the effect of this stress concentration on the fatigue life of the reactor vessel is small and that the rules of ASME Section III are not violated.

In the following discussion, the stresses due to the loads on the shell from the riser braces are not considered. From the "Stress Analysis Brackets" section of the reactor vessel stress report, the stresses are on the order of 1 ksi, which is negligible and will have no appreciable effect on the calculated cumulative usage factor (CUF).

The first step in the analysis is to calculate the stresses at the location of the sample. From the "Top Head and Cylindrical Shell" section of the reactor vessel stress report, it was determined that the range of local membrane plus secondary bending stress at the inside of the vessel at the jet pump riser brace pad location is 22.6 ksi. The report indicated that thermal stresses are not included. Therefore, this stress is due solely to pressure. The report also stated that fatigue requirements were met by demonstrating that the rules of paragraph N-415.1 (Exemption from Fatigue) were satisfied.

Other plant information indicated that the location of interest experiences little fatigue and is exempt from detailed fatigue analysis. Therefore, the fatigue usage at this location (before sample removal) is low, which is consistent with the discussions above.

To complete the stress analysis, a through-wall gradient stress must be determined and the stresses due to the material interface (base metal/cladding) must be accounted for. The relevant parameters are:

Material	A533 Gr B Cl
Thickness	5.00"
Inside radius	103.19"

Since the through-wall gradient stress for heatup and cooldown (HU and CD) was not calculated in the reactor vessel stress report, this was determined using information from the vessel stress report for a plant with similar characteristics. It is reasonable to use the results from the analysis of the similar plant in this assessment since the shells are practically identical for the purpose of calculating the through-wall gradient and the heatup /cooldown rates are the same (100°F/hr).

1

For a linear temperature gradient through a shell, the thermal bending stress is

$E\alpha(\Delta T)/(2)(1-v)$	Equation 3-1
	Equation 2

Where: E = Modulus of elasticity

 α = Coefficient of thermal expansion

 ΔT = Temperature gradient across the wall

 \mathbf{v} = Poisson's ratio = 0.3

It should be noted that the factor 2 in the denominator does not appear in Equation 3-1 when the ΔT used is from mid-wall to the inside surface.

From the reactor vessel stress report, the temperature differences between the inside surface and mid-wall for normal HU and CD are -27.5° F and $+16.5^{\circ}$ F, respectively. For the subject reactor vessel base material listed above at 550°F, E = 26.7E6 psi and $\alpha = 7.77E-6/^{\circ}$ F. Using $\Delta T = [(16.5) - (-27.5)] = 44.0^{\circ}$ F in Equation 3-1 (with the factor of 2 removed in the denominator since the ΔT used is from mid-wall to the inside surface), the stress range due to normal HU and CD is 13.1 ksi.

Finally, the effect of the difference in thermal expansion between base metal and cladding must be included. The cladding/base metal interface stress is due to differences in the coefficient of thermal expansion only, as the interface is at the same temperature. The equation for this interface stress is

 $E(\Delta \alpha)(\Delta T)/(1-\nu)$ Equation 3-2

Where: E = Modulus of elasticity

 $\Delta \alpha$ = Difference in material coefficients of thermal expansion

 ΔT = Material temperature – stress free temperature

 \mathbf{v} = Poisson's ratio = 0.3

For 304 stainless steel (used for conservatism versus a nickel-based weld metal) at 550°F, E = 25.6E6 psi and α = 9.45E-6 /°F, the corresponding difference between the cladding and low alloy base metal coefficients of thermal expansion is $\Delta \alpha$ = [(9.45E-6)-(7.77E-6)] = 1.68E-6 /°F. With a stress free temperature of 70°F, ΔT = (550-70) = 480°F. Therefore, using Equation 3-2 with E = 25.6E6 psi, the interface stress is 29.5 ksi.

Next, the stress concentration factor needs to be determined. This will be based on Reference 4, Fig. 6.1 (c). Using R = 0.13", h = 0.10", D/d = 1, a stress concentration factor of 2.8 is obtained. It should be noted that using h = 0.10 adds additional conservatism for slight variations in the depth of the sample (nominally 0.06" max) due to the weld pad surface variations that may be expected in the field.

To enter the ASME Section III design fatigue curve (1965 Edition), the alternating, i.e., (half-range) stress intensity is formed by multiplying the product of stress range (22.6 + 13.1 + 29.5 = 65.2 ksi) and stress concentration factor (2.8) by one-half. This yields a value of 91 ksi. From ASME Section III, Fig. N-415 (B), the allowable number of cycles is 1500.

The discussion in the Monitoring and Equipment Cycles report indicates that different parts of the vessel were analyzed for different numbers of heatup/cooldown cycles. A value of 300 cycles envelopes the range of values identified in the Monitoring and Equipment Cycles report, including those required for power re-rate. Of course, some of these cycles have already been used but we will use the value of 300 cycles for conservatism. The cumulative usage factor (CUF) is then CUF=300/1500=0.2. This is well below the allowable value of 1.0 and, as such, the formation of a fatigue crack is not expected.

3.2.2 Material Evaluation

The BWR JPRB weld pad is fabricated from Type 308L/309L weld metal. Stainless steel (e.g., Type 308L) weld metal has been found to be very resistant to intergranular stress corrosion crack (IGSCC) initiation in a BWR environment and has been used to provide a protective cladding on the recirculation piping inside surface (Reference 5). However, these materials may be susceptible to IGSCC should a heavily cold worked surface or a metallurgical crevice be present (Reference 6). A metallurgical crevice is defined as a lack of fusion, a visible crack, or smeared metal.

3.2.2.1 Cold Work

Numerous evaluations of IGSCC in stainless steel materials in BWRs (References 7 and 8) have shown that cold work increases the susceptibility to cracking of plate and piping stainless steel materials (e.g., Type 304/304L). However, the duplex microstructure (i.e., austenite and ferrite) of these weld metals makes them significantly more resistant to IGSCC (Reference 5).

If cold work were to introduce an initiating defect or crack on the surface of the Type 308L/309L stainless steel weld metal, crack growth would be very limited or non-existent due to the combination of low carbon and significant ferrite in the weld metal. Laboratory studies and examination of field components have provided evidence that austenitic stainless steel weld metals are essentially immune to IGSCC when they contain ferrite levels typical of those present in 308L/309L material (Reference 9).

There are many locations throughout the system where Type 308L/309L weld metal has been ground during manufacturing, thereby introducing cold work to the surface, and there has been no evidence to date of IGSCC. Therefore, the small amount of cold work resulting from the sampling process, which would be significantly less than that introduced in other BWR components during original construction, is considered acceptable.

3.2.2.2 Crevices

Type 308L/309L stainless steel weld material is susceptible to IGSCC in a crevice situation (References 8, 10, and 11). Therefore, a metallurgical crevice cannot be left in place. In the absence of crevices, there is little experience or information that suggests cracks would initiate in these pad materials. The process qualification (see Section 3.1.2) will show that no crevices (PT exam and visual inspection consistent with BWRVIP requirements, i.e. EVT-1) have been created during the sample removal process. Therefore, IGSCC initiation, resulting from the introduction of a crevice during the sample removal process, is not an issue.

3.2.2.3 Metallurgical Examination

Metallurgical examinations were performed on the jet pump riser brace weld pad qualification samples. The samples were examined for evidence of laps, tears, or crevices, and none were found. The samples were also examined for excessive cold work, and none was found.

Two samples used by FTI in the performance qualification of the jet pump riser brace sampling tool were analyzed. The samples were identified as RBW-001 and RBW-002. A sample which encompassed a machining divot was removed from each sample, see Figures 3-6 and 3-7. The sample from RBW-001 was identified as 1-2 and the sample removed from RBW-002 was identified as 2-3.

Each sample was cross-sectioned through the center line of the divot, mounted in phenolic resin and polished according to accepted metallurgical practices. The cross sections were examined in the un-etched and etched conditions at 50X and 100X. A typical etched cross section of sample 2-3 is shown in Figures 3-8 and 3-9.

The examination revealed a dendritic grain structure with small nonmetallic inclusions. This structure is typical of stainless steel weld metal deposits. Both cross sections had a very distinct edge with no evidence of laps, tears or crevices. There was very little deformation of the dendritic structure indicating minimal cold work had occurred during the sample removal process.

A hardness test was performed on the surface of the weld metal build up on sample RBW-002. This sample was adjacent to divot 7, see Figure 3-6, and had a Rockwell hardness of 16.4C (~ 96Rb). A hardness test was performed at the bottom of divot 7, see Figure 3-6, and had a Rockwell hardness of 15.4C (~ 95Rb). This indicates that the sample removal process does not induce excessive cold work in to the weld metal.

Based upon the above examinations and tests performed on samples RWB-001 and RWB-002, it would appear that the machining process proposed for removal of samples from the jet pump riser pads is acceptable for use. The machining process did not create laps, tears or crevices and did not introduce excessive cold work into the weld metal pad. Based upon the examinations and tests, it would appear that the divots in the weld pad resulting from the removal of samples should not produce an initiation site for stress corrosion cracking.

3.3 Sample Analysis Results

All samples removed in the BWRVIP/NRC program were analyzed for helium content, initial boron content and fast and thermal fluence. Detailed results for all three plants are presented in Reference 1.

No information from the plant operator is generally required for the helium and boron determinations. Correcting the measured as-received boron contents back to their original conditions prior to irradiation requires an estimate of the plant thermal fluence exposures. Usually these corrections are small (<1%), and therefore large uncertainties in these estimates can be tolerated.

Additional information is usually required for determining the plant fluences. As a minimum, this includes the end of irradiation date and a reactor power time history. With this information, fluences can be determined using a generic BWR neutron spectrum. If more accurate spectra are available, this will reduce the overall uncertainties in the fluence determinations to some extent. For one plant, additional information on the fuel pin power distribution was also required to improve the uncertainties in the measured fluence values. The detailed results for this plant's analyses are attached in Appendix A to show the types of results that can be expected from the analysis laboratory and to show the additional information that was required for this one case.



Figure 3-1 Typical BWR Jet Pump Configuration



Figure 3-2 Typical Riser Brace/Pad Configuration

BWRVIP/NRC Jet Pump Riser Brace Pad Sampling



Figure 3-3 Jet Pump Riser Brace Pad Sampling Tool



Figure 3-4 Full Height Mockup Tower



SECTION Z-Z 3X





TYPICAL CONFIGURATION FOR MONTICELLO

TYPICAL CONFIGURATION FOR HATCH & RIVERBEND

Figure 3-5 Proposed As-Left Riser Brace Pad Configuration

BWRVIP/NRC Jet Pump Riser Brace Pad Sampling



Figure 3-6 RBW-002 Cut line layout for Sample 2-3



Figure 3-7 RBW-001 Cut line layout for Sample 1-2



Figure 3-8 Cross Section of Sample 2-3 (100X)



Figure 3-9 Cross Section of Sample 2-3 (100X)

4 REFERENCES

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A EXAMPLE OF ANALYTICAL RESULTS

Presented below are the results of the helium, boron and fluence analyses for one of the three plants that were sampled as part of the BWRVIP/NRC project. The results are presented essentially as received from the laboratory. The results were originally reported by B. M. Oliver and L. R. Greenwood of PNNL.

A.1 Helium and Boron Analyses

A.1.1 Summary

Results of helium and boron analyses on four jet pump riser brace (JPRB) weld pad samples provided by FRA-ANP are reported.

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Estimated uncertainty in the boron concentrations ranged from $\sim 3\%$ to $\sim 6\%$. Uncertainties greater than about 2% are attributed to heterogeneities in the boron contents in the material for the milligram and sub-milligram sized samples analyzed. Overall, however, the range of heterogeneity is very similar to that observed in previous sets of reactor steel. The range of boron concentration values, however, is higher than can likely be explained by measurement uncertainty or boron heterogeneity, and suggests real variations in the boron contents between the four JPRB samples.

A.1.2 Analysis Samples

Four sets of stainless steel samples were received from FRA-ANP on February 1, 2000 for helium, boron, and activation analysis. All of the samples had been taken remotely from jet pump riser weld pads from a commercial reactor between January 15 and January 18, 2000, and were individually contained in stainless steel filter assemblies. Each of the samples was in the form of multiple small machine chips.

Sample 1 was intermingled with rust and organic matter from the flow water supply. This material had essentially no activity, and was separated out prior to subsequent sample preparation. Content Deleted -

EPRI Proprietary Information

Content Deleted -EPRI Proprietary Information

A.1.3 Sample Preparation

Samples for helium and boron analyses were taken from a subset of the as-received material. For the initial helium analyses, single discrete metal pieces were selected. The pieces were chosen with the aid of a low-power stereo microscope to be as representative of the each sample set as possible. Pieces that were potentially oxidized, or that had other features not representative of the lot as a whole, were not used for the analyses.

For the boron determinations, multiple chips were selected as described above and wrapped in aluminum foil for neutron exposure at the University of Missouri Research Reactor (MURR). Following the MURR exposure, single specimens were selected from the set for additional helium analysis to determine the boron content. Details of the neutron exposure are given in Section A.1.4.

Prior to analysis, each specimen was cleaned in acetone, air-dried, and then weighed using a microbalance with calibration traceable to the National Institute for Standards and Technology (NIST). Specimen masses ranged from ~0.2 to ~2 mg each. Mass uncertainty is conservatively estimated to be ± 0.002 mg.

A.1.4 Neutron Exposure

Determination of the boron content in the samples was made by first irradiating a selected subset of the sample material in a thermalized neutron field in the MURR and then measuring the increase in the sample helium content. In a thermalized neutron field, helium generation will be predominately through (n,α) reactions with ⁶Li or ¹⁰B, or with Ni through the two-stage reaction ^{5*}Ni (n,γ) ⁵⁹Ni (n,α) . Lithium is assumed to be an improbable impurity in steels due to its relatively high volatility. If the materials have already been exposed in a neutron environment, as was the case here, corrections are also applied to determine the original level of boron in the material.

The boron characterization samples and the dosimetry samples were packaged together inside an aluminum rabbit assembly originally supplied by Brookhaven National Laboratory (BNL). The samples were contained inside a small aluminum capsule, wrapped with aluminum foil, and held in the approximate axial center of the rabbit. The aluminum rabbit, capsule, and foil were fabricated from Type 1100 aluminum.

Figure A-1 shows a diagram of the sample loading arrangement inside the rabbit. The rabbit was backfilled with argon, and sealed prior to shipment to MURR. Table A-1 lists the individual samples in the irradiation capsule. The aluminum rabbit was irradiated at MURR in a reflector location for 24 hours. Details of the irradiation are given in Table A-2.

Multiple samples of Al-Li alloy wire (Reference 12) were included in the irradiation capsule in order to characterize the thermal neutron fluence. This alloy was originally fabricated by the Central Bureau for Nuclear Measurements (CBNM) in Belgium specifically for neutron

dosimetry applications. The stated CBNM composition of the Al-Li alloy is 0.705 ± 0.025 wt.% Li, with a lithium isotopic composition of 95.82 ± 0.08 at.% ⁶Li. Independent analysis at Argonne National Laboratory showed a lithium content of 0.730 ± 0.004 wt.%, and an isotopic composition of 95.67 ± 0.04 at.% ⁶Li; essentially in agreement with the CBNM values but with lower uncertainties. For analysis purposes, a composition of Al-0.73±0.01 wt.% Li, with a ⁶Li content of 95.7 ± 0.1 at.% is assumed.

Also included as a control material for the boron determinations was a sample of PNNL's Lot 7 vanadium material. This material is used for the fabrication of Helium Accumulation Fluence Monitor (HAFM) capsules and radiometric monitors, and has been characterized for boron content in a variety of irradiation environments yielding a current average value of 4.5 ± 0.2 wt. ppm.

A.1.5 Helium Analysis Procedure

Helium analyses on all specimens were conducted by isotope-dilution gas mass spectrometry following vaporization in a resistance-heated tungsten-wire or graphite crucible in one of the mass spectrometer system's high-temperature vacuum furnaces. (Reference 13). Duplicate helium analyses are routinely performed to give an indication of the analysis reproducibility and to give an indication of the helium homogeneity in each sample. This was important for the present work, where heterogeneity in the boron impurity was a concern, particularly for Samples -1 and -3, where limited sample material was available. To help clarify the heterogeneity question, triplicate analyses were conducted on selected samples as indicated in Table A-3.

The absolute amount of ⁴He released was measured relative to a known quantity of added ³He "spike." The ³He spikes were obtained by expanding and partitioning known quantities of gas through a succession of calibrated volumes. (Reference 14). The mass spectrometer was calibrated for mass sensitivity during each series of runs by analyzing known mixtures of ³He and ⁴He, as specified in PNNL Procedure RPG-MSL-1000.

A.1.6 Helium Analysis Results

The results of the helium measurements are given in Table A-3 and A-4. Table A-3 gives the results of the helium analyses on the Al-Li alloy dosimetry material. Table A-4 gives the helium concentrations measured in the characterization samples both before and after neutron exposure at MURR.

Helium concentrations were measured in three of the four Al-Li alloy wires included in the irradiation. These concentrations are given in Table A-4, and are listed as total atoms of helium released, and as helium concentrations in atomic parts per million (10^6 atom fraction) relative to the total number of ⁶Li atoms in each specimen. Conversion from total helium to helium concentration was based on a calculated number of ⁶Li atoms per gram of 0.06942 x 10^{22} .

Helium concentrations in the Al-Li alloy were used to determine the thermal neutron exposure in MURR. From Table A-3, the mean helium concentration (in terms of atom fraction) is 0.005048 ± 0.000079 . Correcting for burnup of the ⁶Li isotope and for neutron self-shielding yields a

value of 0.005370 ± 0.000084 , which represents the ⁶Li reaction probability $\sigma \phi t$. Burnup corrections were made using the standard exponential formulation. Self-shielding corrections were based on transport theory approximations from Case et al. (Reference 15), and were 6.1%. Uncertainty in the self-shielding correction is conservatively estimated to be ~25% of the correction factor. Assuming a 2200 m/s ⁶Li(n, α)³H neutron cross section of 941 barns, yields an effective thermal neutron fluence of 5.71 x 10¹⁸ n/cm². It should be noted, that this value is in excellent agreement with an independent determination of 5.75 x 10¹⁸ n/cm² obtained from the gamma analysis data used to determine the plant fluences.

Helium concentrations measured in the samples, both before and after neutron exposure at MURR, are given in Table A-4. The results are listed as total atoms of helium released and as helium concentrations in atomic parts per billion (10^{-9} atom fraction). Residual (pre-exposure) helium concentration in the PNNL vanadium has been determined previously to be <0.03 appb. Conversion from total helium to helium concentration was based on the following calculated values:

308L SS	1.09 x 10 ²² atoms/gram
Vanadium	1.182 x 10 ²² atoms/gram

The value for the 308L SS material is a nominal value for Type 308L stainless steel. It should be noted, however, that these numbers, and the helium concentrations obtained using it, are not very sensitive to small changes in material composition.

Absolute uncertainty in the measured helium concentrations is estimated to be ~2% (1 σ). This uncertainty results from the cumulative uncertainties in the sample mass, the isotope ratio measurement, and the spike size.

A.1.7 Boron Determinations

Calculated boron contents in the samples are given in Table A-5. Column 6 gives the calculated helium contribution from the nickel component in the as-received material, accounting for burnup of ⁵⁸Ni and ingrowth of ⁵⁹Ni from the material's prior reactor exposure. Cross sections used for the nickel two-stage reaction were those determined by Greenwood et. al. (Reference 16). The nickel contributions in Table A-5 are essentially negligible at ~0.1 appb. A conservative error estimate of 10% is assigned to this value. Helium contributions from threshold neutron reactions (greater than ~2 MeV) were calculated to be <1 %, and were neglected.

The Column 7 values in Table A-5 represent the calculated ¹⁰B content in the as-received samples *prior* to the MURR exposure. These values were calculated from the "net" increase in the helium content attributable to the boron, obtained by subtracting the values in Columns 4 and 6 from those in Column 5, and assuming a 2200 m/s cross-section for the ¹⁰B(n, α) reaction of 3838 barns. Burnup of ¹⁰B during the MURR irradiation, which amounted to 2.2 %, was accounted for in the calculations.

Columns 8 and 9 gives an estimate of the original (unirradiated) natural boron content in each of the samples, and the estimate overall uncertainty. These values were determined from the calculated as-received ¹⁰B contents in Column 7, corrected for burnup of ¹⁰B during the previous plant exposure, and assuming a ¹⁰B isotopic content of 19.9 atom %. Calculated burnup levels for the previous plant exposure, based on the thermal neutron fluences in Column 2 calculated at PNNL from the radiometric analyses, ranged from ~0.3 % to ~0.7 %.

A.1.8 Uncertainty in Boron Determinations

Error sources in the determination of the boron contents are given in Table A-6. The largest error source is the variability in the measured pre- and post-irradiation helium concentrations. Variability in the measured helium concentrations is attributed largely to heterogeneity in the boron contents for the milligram and sub-milligram sample masses analyzed. Total uncertainty estimates, determined from the quadrature sum of the various uncertainty components, are given in the last column of Table A-6.

Uncertainty in the nickel content of the stainless steel material is given as ± 0.5 wt. % based on the x-ray fluorescence analyses performed as part of the radiometric analyses. Uncertainty in the calculated plant thermal neutron fluence values was arbitrarily assigned as ± 10 %. For the present measurements, however, uncertainty in the plant thermal fluence has a negligible effect on the final calculated boron contents.

A.1.9 Discussion of Results

A.2 Retrospective Neutron Dosimetry

A.2.1 Summary

Four scraping samples were obtained from the jet pump riser brace pads at positions 1, 3, 4, and 5 around the pressure vessel of the reactor and delivered to PNNL for analysis. Each sample consisted of multiple, small stainless steel turnings. Sample 1 showed some contamination from residual material in the sampler. This residual material had little contamination and did not appear to affect our analyses. Selected subsamples from each group of turnings were initially gamma counted to determine the residual activation products. Subsamples were also analyzed for the beta-emitters ³⁵Fe and ⁶³Ni. The chemical composition of each steel sample was measured by x-ray fluorescence. All of the measured activities were then converted to saturated activation rates by taking into consideration the reactor power history. Thermal and fast neutron fluences were finally determined at each position using spectral-averaged cross sections determined for a typical BWR pressure vessel spectrum. The neutron fluences were first determined using the total reactor power history. However, this analysis showed that shorter-lived reactions predicted lower neutron fluences than the longer-lived reactions. This effect appears to be readily explained by changes to the reactor core configuration that were made to reduce neutron leakage, thereby reducing the neutron fluxes at the locations that were sampled. The analyses and results are detailed below.

A.2.2 Gamma Energy Analysis

Selected subsamples were weighed and then gamma counted in duplicate using high efficiency intrinsic germanium detectors following procedure PNL-ALO-450. The detectors are calibrated relative to NIST and other accepted standards, and control counts are performed daily to ensure continuing energy and efficiency calibrations. All four of the samples showed the presence of ⁵¹Cr, ⁵⁴Mn, ⁵⁷Co, ⁵⁸Co, ⁵⁹Fe, ⁶⁰Co, and ⁶⁵Zn. The measured activities and total propagated uncertainties in microCuries/milligram are listed in Table A-7. All of the activities were corrected for decay to the date of the reactor shutdown at the end of cycle 19 on January 6, 2000. The samples were removed from the reactor between January 15 and 18, 2000; however, it is assumed that they did not see any significant neutron exposure following the reactor shutdown.

The ⁵¹Cr, ⁵⁹Fe, ⁶⁰Co, and ⁶⁵Zn activities are due to thermal neutron activation of Cr, Fe, and Co, Cu, and Zn impurities in the samples. The other activities are produced by the fast neutron reactions ⁵⁴Fe(n,p)⁵⁴Mn, ⁵⁸Ni(n,p)⁵⁸Co, and ⁵⁸Ni(n,2n+np)⁵⁷Co. Multiple counts on independent subsamples were averaged to determine the values in Table A-8. The uncertainties include both the counting statistics and scatter in the duplicate counts. The ⁶⁰Co value for position 1 showed considerable scatter, possibly due to the contamination of this sample by the residual material in the sampling mechanism. ⁶⁰Co is also produced by two other reactions, namely ⁶⁰Ni(n,p)⁶⁰Co and from the decay of ⁵⁹Fe to ⁵⁹Co. However, calculations indicate that both of these reactions make a negligible (< 0.7%) contribution to the ⁶⁰Co that is produced from the ⁵⁹Co impurity in the steel.

A.2.3 ⁵⁵Fe Measurements

Subsamples measuring about 1.5 mg were dissolved in heated HCl and brought to a known volume in distilled water. Aliquots were taken and spiked with 2-mg Fe carrier to be used as a yield monitor. The iron fraction was purified using extraction chromatography following procedure PNL-ALO-435. The sample matrix was converted to 8M HNO, and loaded onto a TRU Resin column. Iron was retained on the column, impurities were eluted with 8M HNO₃. The iron was then eluted with 2M HNO₃. An Fe(OH), precipitate was then formed with stirring by adding concentrated NH₄OH. The Fe(OH), precipitate was loaded onto a Tuffryn® filter, dried, and covered with 6-micron mylar film. The filters were then counted using thin-window germanium detectors (Low Energy Photon Spectrometers or LEPS). Prior tests have demonstrated that x-ray fluorescence is not significant with this mounting scheme. The LEPS detectors were calibrated using "Fe NIST standards prepared in identical geometries. The sample activities were corrected for decay and x-ray self-absorption was calculated to be negligible. After counting, the Fe(OH), was dissolved in a known volume of 3M HNO, and an aliquot analyzed by Inductively Coupled Plasma Atomic Emission Spectroscopy (ICP-AES) to determine Fe concentration and thus radiochemical yield. The radiochemical yields averaged 95%. Sample duplicates showed good repeatability and the blank spike and matrix spike averaged 95% yield-corrected recovery. The "Fe activities and uncertainties are listed in Table A-7. The ⁵⁵Fe activity is produced by the ⁵⁴Fe(n, γ) thermal neutron reaction.

A.2.4 ⁶³Ni Measurements

Subsamples measuring about 1.5 mg were dissolved in HCl and brought to a known volume in distilled water. Aliquots were taken and spiked with 2-mg Ni carrier to be used as a yield monitor. After a matrix adjustment, the nickel fraction was purified using an on-column nickel dimethyglyoxime precipitation reaction following procedure PNL-ALO-495. Nickel was retained on the column; impurities were eluted with a basic ammonium citrate solution. The nickel was then eluted with 3M HNO3. The strip solution was carefully evaporated to near dryness, brought to a known volume with 0.1M HCl, and split for radiochemical recovery determination by ICP-AES and ⁶³Ni analysis by liquid scintillation counting following procedure PNL-ALO-474. The chemical yields averaged 99%. A minor correction for ⁶⁰Co tailing into the ⁶³Ni region was made. Calibrations were performed using ⁶³Ni standards obtained from NIST. Sample duplicates showed good repeatability and the blank spike and matrix spike averaged 101% yield-corrected recovery. The decay-corrected ⁶³Ni activities are listed in Table A-7. The ⁶⁴Ni activity is produced by the ⁶²Ni(n, γ) thermal neutron reaction.

A.2.5 Calculation of Saturated Activation Rates

Table A-7 lists the measured activities and uncertainties corrected to the end of irradiation time for each sample and reaction. Nuclear decay data were taken from the Table of Radioactive Isotopes, E. Browne, R. Firestone, and V. Shirley, Wiley, 1986. The activities were converted to saturated activation rates by correcting for the decay during irradiation, atomic weight, elemental abundance, isotopic abundance, gamma self-absorption, neutron self-absorption, and nuclear burnup. These factors are shown in Table A-8 and are discussed below.

The decay during irradiation was determined using the BCF computer code that integrates the decay of each isotope for each period of reactor operation and downtime. Reactor time history as provided by the client for the reactor, is shown in Figure A-2. The reactor operated from 7/06/71 to 1/06/00 for a total exposure of 7456 FPD (full power days) at a power level of 1775 MWt. It should be noted that the reactor operated at a power of 1670 MWt over most of its history. The reactor was rerated to 1775 MWt during the course of cycle 19. The saturated activities were normalized to the new power level of 1775 MWt. Values can be renormalized for the lower power level of 1670 MWt by simply multiplying by the power ratio. (In this case, the total exposure should be stated as 7925 FPD at 1670 MWt). The accuracy of the reactor power history correction factors depend on the halflife of each isotope relative to the details of the irradiation histories. The longer-lived isotopes, such as ⁶³Ni truly integrate over the entire irradiation history. However, shorter-lived isotopes, such as ⁵⁹Fe (45 day) or ⁵⁸Co (71 day) are only dependent on the last year or so of reactor operation and the decay corrections are thus more uncertain since they depend critically on the fine details of the irradiation history. For this reason, neutron fluence results are more accurate for the longer-lived isotopes. In an attempt to improve the reliability of the shorter-lived isotopes, fine details of the power history from cycle 19 were used in the calculations. For the other cycles, the average power level was used for each cycle, as shown in Figure A-2.

Estimated gamma self-absorption corrections varied from 1-2% for the direct counting of the turnings. As mentioned above, there were no significant x-ray absorption or fluorescence effects for ⁵⁵Fe. Neutron self-absorption corrections were not performed since we do not have knowledge of the geometry of the individual turnings during reactor operation. It is assumed that the samples were removed from the surface of the pressure vessel. In this case, the neutron self-absorption corrections would be negligible for stainless steel. Due to the relatively low neutron fluence levels, nuclear burnup and transmutation corrections for both target and product isotopes were negligible.

Examination of the neutron fluences derived later from the data in Table A-9 showed a trend with the halflife of the reaction product. This effect was most likely due to efforts by the utility to reduce the neutron leakage in order to improve plant efficiency. The net effect of these changes was that the neutron flux at a specified position on the JPRB's might not depend linearly on the reactor thermal power. Corrections to account for these changes are discussed below.

A.2.6 X-Ray Fluorescence Measurements

The steel samples have the nominal composition of Type-308 stainless steel. In order to determine the exact composition of each sample more accurately, subsamples were analyzed by energy dispersive x-ray fluorescence (EDXRF) techniques at KLM Analytical in Richland, WA. In this procedure, samples are bombarded with an electron beam and the x-rays are measured with a high-resolution detector. Since the samples were radioactive, x-ray spectra were also taken with the beam off and this background was subtracted from each EDXRF x-ray spectrum. NIST 304 stainless steel standards were analyzed at the same time to calibrate and verify the performance of the equipment and interelement corrections. The results of the x-ray measurements are listed in Table A-9.

The samples were also analyzed by wavelength dispersive x-ray fluorescence to determine the level of the Co impurity. The results were inconclusive for samples 1 and 3 due to the small amount of available material in these samples. However, good results were obtained for samples 4 and 5. We also calculated the Co impurity concentration from the boron measurement in the Missouri University Research Reactor, as described in the accompanying helium and boron report. The Co concentration can be directly calculated from a gamma energy analysis of the irradiated samples using a neutron activation technique, including subtraction of the initial ⁶⁰Co activity. These results are also shown on Table A-9. The Co concentrations for positions 4 and 5 are in excellent agreement with the WDXRF measurements so that we have confidence in these values for positions 1 and 3 where the WDXRF values have high uncertainties.

A.2.7 Neutron Fluence Evaluations

Saturated activities are equal to the integral over neutron energy of the neutron activation cross section times the neutron flux spectrum. In principle, the neutron flux spectra can be determined using neutronics calculations; however, such information was not provided. An estimate of the thermal neutron fluence can be obtained by dividing the saturated activities by the 2200 m/s thermal neutron cross section and multiplying by the total irradiation times using cross section data obtained from the National Nuclear Data Center at Brookhaven National Laboratory (values are listed in Table A-8). However, a better estimate of the thermal neutron fluence can be obtained by correcting for the presence of epithermal neutrons. A simple way to do this is to set each saturated activity equal to the sum of the thermal flux times the thermal cross section plus an epithermal flux times the resonance integral. This correction is also listed in Table A-8 and is shown as a corrected thermal neutron cross section. More accurate estimates of the thermal and epithermal fluences require a calculation of the neutron flux spectrum at each position rather than using a generic BWR neutron spectrum. For the present analyses, a neutron energy spectrum obtained previously from General Electric for a surveillance capsule position for a GE BWR reactor was used.

The neutron fluences from the calculated saturated activities and cross sections are listed in Table A-10. As was discussed above, the values from the various thermal neutron reactions show a trend with halflife. Due to the short halflifes of ⁵⁹Fe and ⁵¹Cr, results from these reactions are not as reliable as those from the other reactions and the thermal fluences were determined by averaging fluences from the longer-lived reactions. The standard deviations on the three thermal neutron measurements range from 8 to 17%.

The gamma counting data can also be used to estimate the fast neutron fluence. In order to accurately determine the fast fluence above 1 MeV, or any other energy threshold, it is necessary to know the energy dependence of the neutron flux spectrum. These spectral-averaged activation cross sections were calculated from the GE BWR neutron spectrum mentioned above and they are listed in Table A-8. The standard deviations in the averages of the two fast neutron reactions range from 13 to 30%. However, absolute uncertainties may be somewhat larger due to the uncertainty in the choice of the spectral-averaged cross sections. The fast fluences derived from ⁵⁴Mn (312 d) are consistently higher than those from ⁵⁸Co (71 d) due to the flux non-linearity problem with the reactor power history, as discussed above. The difference between the ⁵⁴Mn and ⁵⁸Co derived fast fluences also depends on the spectral-averaged cross sections. As was the case for the thermal and epithermal fluences discussed above, more accurate determinations

would require detailed neutron flux spectra calculations for each of the four JPRB positions. The fluence ratios from Fe/Ni are very consistent at about 1.21 for positions 1, 4, and 5. However, the Fe/Ni ratio at position 3 is 1.54. This was checked by triplicate gamma counts of separate turnings from sample 3 and all three results agreed within $\pm 1\%$. This may be due to a difference in the neutron energy spectrum at this location. Table A-10 lists the fast neutron fluences for thresholds of 0.1 and 1.0 MeV at each of the four positions. These values were calculated by averaging the Fe and Ni results.

A.2.8 Edge Fuel Bundle Power History Corrections

As was discussed above, the various thermal neutron fluence estimates from different reactions appear to show a trend with the reaction product halflife, indicative of a potential problem with the reactor power history corrections for each reaction. Additional information was received from the plant operator indicating that changes had been made to the reactor fuel configuration in order to reduce the neutron leakage, thereby lowering the flux on the JPRB positions. The average edge power peaking factor for the reactor ran at about 60% of the core average for the first 11 fuel cycles, dropping to about 40% for the last 7 fuel cycles. These factors were used to determine a core edge power history, as shown in Figure A-4. This modified reactor power history was then used to determine revised saturation factors for each of the reactions. The resultant neutron fluence values are shown in Table A-11. The revised saturated activity values are given in Table A-12. As can be seen, the previous trend with reaction product halflife is virtually eliminated and all of the thermal reactions give consistent results with much lower standard deviations for the average fluences than shown previously in Table A-10. It is important to note that the thermal neutron fluences derived from the longest-lived reactions are not affected very much by these changes and that the new average thermal fluences are in excellent agreement with the longest-lived values.

The fast neutron fluences are derived from two reactions that are both relatively short-lived (312 day and 71 day). Hence, using the revised core edge power history significantly changes both reaction rates. A comparison of Tables A-10 and A-11 shows that the fast neutron fluences are about 30% higher using the core edge power history. This difference is to be expected since the total reactor power history generally shows increased power as a function of time whereas the core edge power history shows a distinct drop around cycle 12 due to the efforts to reduce neutron leakage. Hence, the reaction correction factors for shorter halflives are about 30% higher for the core edge power history, as was seen with the thermal neutron reactions.

Position 3 appears to show more scatter in the neutron fluences than the other positions for reasons that are not understood. Multiple gamma counts from separate subsamples showed excellent agreement, confirming that the differences are real and not due to sample inhomogeniety or contamination.

A.2.9 Conclusions

The thermal and fast neutron fluences have been determined with uncertainties ranging from 6 to 29% at each of the four JPRB positions. The data clearly show the effects of the decrease in the core edge neutron fluxes caused by the changes in the core configuration to reduce the neutron leakage and the fluences in Table A-11 are thus recommended. The absolute accuracy of the fast neutron fluences might be significantly improved if neutron spectral calculations were performed at each of the four JPRB positions, taking into account the core configurations over the lifetime of the reactor. Such calculations would provide more reliable estimates of the spectral-averaged cross sections for the Fe and Ni reactions, hopefully improving the agreement between these independent measurements of the fast neutron fluences. However, this would require a considerable additional effort that might only provide a marginal improvement in the fluence determinations. The thermal neutron fluences from the different reactions are in good agreement and the absolute accuracy should be close to the standard deviations quoted in Table A-11.



Figure A-1 MURR Irradiation Assembly



Figure A-2 Power History. Cycle 19 is Expanded to Show Fine Detail



Figure A-3 Neutron flux spectrum for a typical GE BWR pressure vessel location



Figure A-4 Reactor Power History using Edge Fuel Bundle Peaking Factors

Sample			Mass	
Name	Material	No.ª	(mg)	Sample Description
SS-1	Stainless steel	1	3.2	Multiple tool chips
SS-3	Stainless steel	1	3.6	Multiple tool chips
SS-4	Stainless steel	1	10.4	Multiple tool chips
SS-5	Stainless steel	1	11.5	Multiple tool chips
AlLi-1 to -4	Al-0.7 wt.% ⁶ Li alloy	4	12.2	Bare wire, 0.5 mm dia. x ~6 mm long
V-1	Vanadium (Lot 7)	1	29.7	Bare wire, 0.9 mm dia. x ~6 mm long

Table A-1Reactor Steel and Dosimetry SamplesIrradiated in MURR

^{*}Number of specimens of sample included in irradiation capsule. The steel samples consisted of multiple chips wrapped in aluminum foil.

Table A-2 Summary of MURR Irradiation Parameters

Parameter	Value
Irradiation Start Date	March 3, 2000
Irradiation Location	H-1 (reflector position)
Irradiation Temperature (°C)	46 (nominal)
Irradiation Time (hours)	24
Calculated Thermal Neutron Fluence (n/cm ²)*	5.71 x 10 ¹⁸
Estimated Fast Neutron Fluence (n/cm ²) ^b	2 x 10 ¹⁷
¹⁰ B Burnup (%)	2.2

"Thermal neutron fluence determined from the measured helium generation in the Al-Li alloy samples (see text).

^bEstimated fast neutron fluence (>1 MeV) in H-1 location.

Table A-3 Helium Concentrations in Al-Li Alloy Dosimetry Samples

			Measured	Helium Co	ncentration
		Mass*	⁴ He	(ap	pm)
Specimen	Material	(mg)	(10 ¹⁵ atoms)	Measured [▶]	Average
AILi-1-A	Al-Li Alloy	0.983	3.505	5136	
AlLi-2-A	66	0.870	3.035	5025	5048 ± 79
AlLi-3-A	14	1.013	3.504	4983	

*Mass of analyzed specimen. Mass uncertainty is ±0.002 mg.

^bHelium concentration in atomic parts per million (10⁶ atom fraction) with respect to the total number of ⁶Li atoms in the specimen.

'Mean and standard deviation (1σ) of analyses.

Table A-4 Measured Helium Concentrations

Table A-5Calculated Boron Contents in Steel Samples

Table A-6 Error Sources for Boron Determinations

Descenter	Estimated Error	Resultant Error in	
Parameter	in Parameter (1 σ)	Boron Content (%) ^a	
Measured:			
He concentrations ^b	0.5 % - 8 %	1-7	
Neutron fluence (MURR irradiation)	2.2%	2.2	
Nickel content (wt. %)	0.5 wt. %	<0.1	
Calculated:			
He generation from nickel component	10%	<0.1	
Prior thermal fluence (n/cm ²)	10%	<0.1	

*Standard error range in calculated boron contents resulting from errors in the corresponding parameter.

*Standard deviations in replicate analyses (pre- and post-exposure combined).

Table A-7 Activity measurements in $\mu\text{CI/MG}$

Reaction	At.Wt	lso.Abn.	Gabs	History	Cross Sections, barns	
Thermal:		<u> </u>			Thermal	Epi.Cor.
⁵⁴ Fe(n, γ)⁵⁵Fe	55.847	0.058	0.998	1.135	2.25	2.30
⁶² Ni(n, γ) ⁶³ Ni	58.69	0.0363	N/A	0.1803	14.5	14.8
⁵⁸ Fe(n, γ) ⁵⁹ Fe	55.847	0.0028	0.993	1.282	1.28	1.35
⁵⁰ Cr(n, γ) ⁵¹ Cr	51.996	0.0435	0.987	1.261	15.9	16.2
⁵⁹ Co(n, γ) ⁶⁰ Co	58.933	1.0	0.993	1.065	37.2	40.4
Fast:					>.1 MeV	>1 MeV
⁵⁴ Fe(n,p) ⁵⁴ Mn	55.847	0.059	0.990	1.202	0.1058	0.1846
⁵⁸ Ni(n,p) ⁵⁸ Co	58.69	0.6808	0.990	1.284	0.1337	0.2333

 Table A-8

 Correction factors and cross sections for each reaction

At.Wt. = atomic weight

Iso. Abn. = isotopic abundance of target (also need elemental abundance in alloy)

Gabs = calculated gamma self-absorption in wires

History = reactor power history correction for decay

Thermal = cross section and fluence derived with 2200 m/s cross section

Epi. Cor. = thermal neutron cross section with epithermal corrections assuming a 53% epithermal flux

 Table A-9

 EDXPF Analyses of the JPRB Steel Samples

Table A-10 Neutron Fluences

TableA-11Revised Neutron Fluences Using Core Edge Power History

 Table A-12

 Saturated Activation Rates in Atom/Atom-Second (Corrected for Core Edge Peak Bundle factors)

Target: Nuclear Power

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