ASSESSMENT OF OPERATIONAL DIFFICULTIES EXPERIENCED IN RECENT HFIR INSTRUMENTED MATERIALS IRRADIATION EXPERIMENTS

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1. INTRODUCTION

Operational difficulties were experienced with some instrumented materials irradiation experiments performed in HFIR during the two-year period preceding the outage for the beryllium change out and reactor upgrades. While none of these problems posed hazards to either the reactor or personnel, some did result in premature termination of experiments, and in two cases resulted in unscheduled shutdowns of the reactor to remove the experiment. This paper will provide a detailed description of the operational problems, the investigations into the cause, and the proposed changes to prevent a recurrence of the problems.

2. DESCRIPTION OF OPERATIONAL DIFFICULTIES

The operational difficulties affected three different irradiation experiments (RB-10J, RB-13J, and RB-14J), and the operation of the junction boxes used to join the gas lines of the experiments to the gas lines of the instrumentation facilities. A schematic of how instrumented experiments are connected to the instrumentation facilities is shown in Figure 1. Typically, experiments are connected to the instrumentation facility during the outage preceding the beginning of irradiation, however, in some cases they may be connected a full cycle before irradiation begins. During every refueling outage all instrumented experiments are removed from the reactor and stored in the in-pool experiment storage facility while remaining connected to the instrumentation facility.

A chronology of events that relate to the operational difficulties is presented in Table 1. The following sections provide detailed information of the difficulties experienced with each experiment, or the junction boxes, roughly in the order of their occurrence.

2.1 RB-10J

The RB-10J experiment was the first HFIR materials irradiation experiment to utilize lithium filled subcapsules. Approval for installation in HFIR required that the lithium be contained within two monitored containments. The primary containment was made of all welded construction with the exception of two braze joints located a few inches above the

top of the primary containment vessel. The braze joints, forming part of the primary containment, were necessary to seal the two thermocouple assembly tubes (TCATs) that went into thermocouple wells in each of the lithium filled subcapsules. Additional braze joints, forming part of the secondary containment, were located in the lower bulkhead and a few inches above the lower bulkhead. All braze joints were completed in accordance with approved ORNL procedures. The arrangement of the subcapsules, the two containments, lower bulkhead, and critical braze joints is shown in Figure 2.

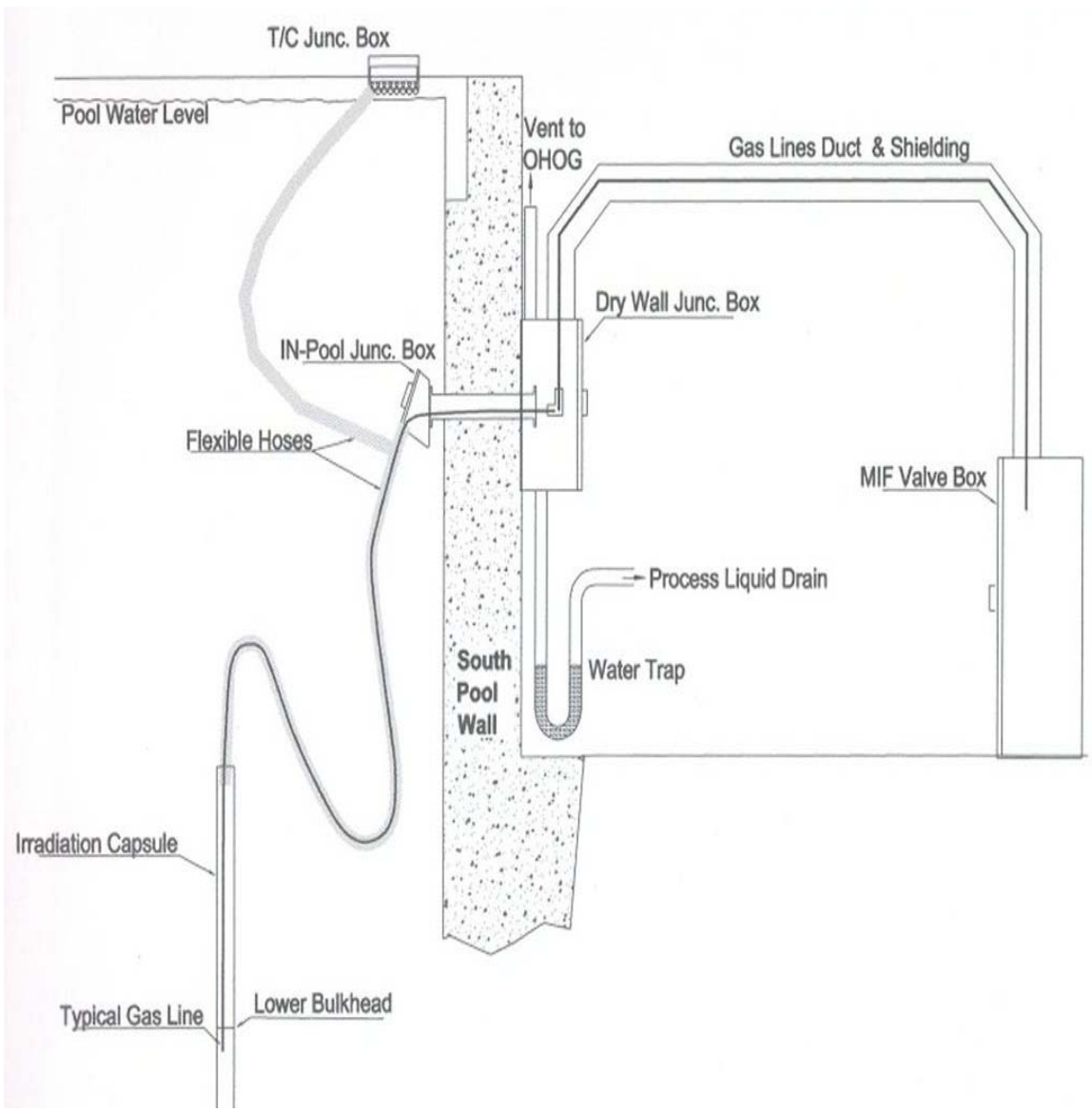


Figure 1. Schematic diagram of instrumented experiment installation

Table 1. Chronology of events relating to operational difficulties experienced with recent instrumented irradiation experiments.

July 22, 1998	Irradiation of RB-13J experiment started
October 13, 1998	Irradiation of RB-10J experiment started
October 23, 1998	A leak in the primary system of RB-10J is determined to be in the part of the primary system that is located within the test region of the capsule. The reactor is shutdown and RB-10J is removed from the reactor, but remains connected to the in-pool junction box
March 15, 1999	Irradiation of TRIST-TC1 experiment started
May 26, 1999	After reinstallation following the EOC 369 refueling outage RB-13J develops a leak making it impossible to maintain pressure in the capsule.
May 28, 1999	While developing an alternative mode of operation for RB-13J water is inadvertently permitted to partially fill the in-pool and dry-wall junction boxes. Experiments connected to the in-pool junction box include RB-10J, RB-13J, RB-14J, TRIST-TC1
June 3, 1999	RB-14J experiment started. RB-13J is not reinstalled in reactor, terminating the experiment after 8 of 10 planned cycles of irradiation.
June 27, 1999	TRIST-TC1 experiment is successfully completed
August 2, 1999	RB-13J and TRIST-TC1 are disconnected from facility
August 11, 1999	RB-10J experiment is re-started under a modified mode of operation
August 27, 1999	There are indications that a leak has developed in the secondary containment system of RB-10J. While the experiment is still operating within the approved conditions for operation, an administrative decision is made to shut down and remove the experiment. An administrative decision was also made to remove RB-14J.
January 27, 2000	Irradiation of RB-14J was resumed
May 12, 2000	RB-13J shipped to hot cells
June 6, 2000	After continuous increase in unexplainable moisture indications of two moisture monitors on the effluent of RB-14J, it was decided to terminate the irradiation after 7 of 10 planned cycles of irradiation
July 26, 2000	TRIST-TC1 shipped to hot cells
October 6, 2000	RB-10J and RB-14J are disconnected from facility
November 7, 2000	RB-14J shipped to hot cells
June 5, 2001	RB-10J shipped to hot cells

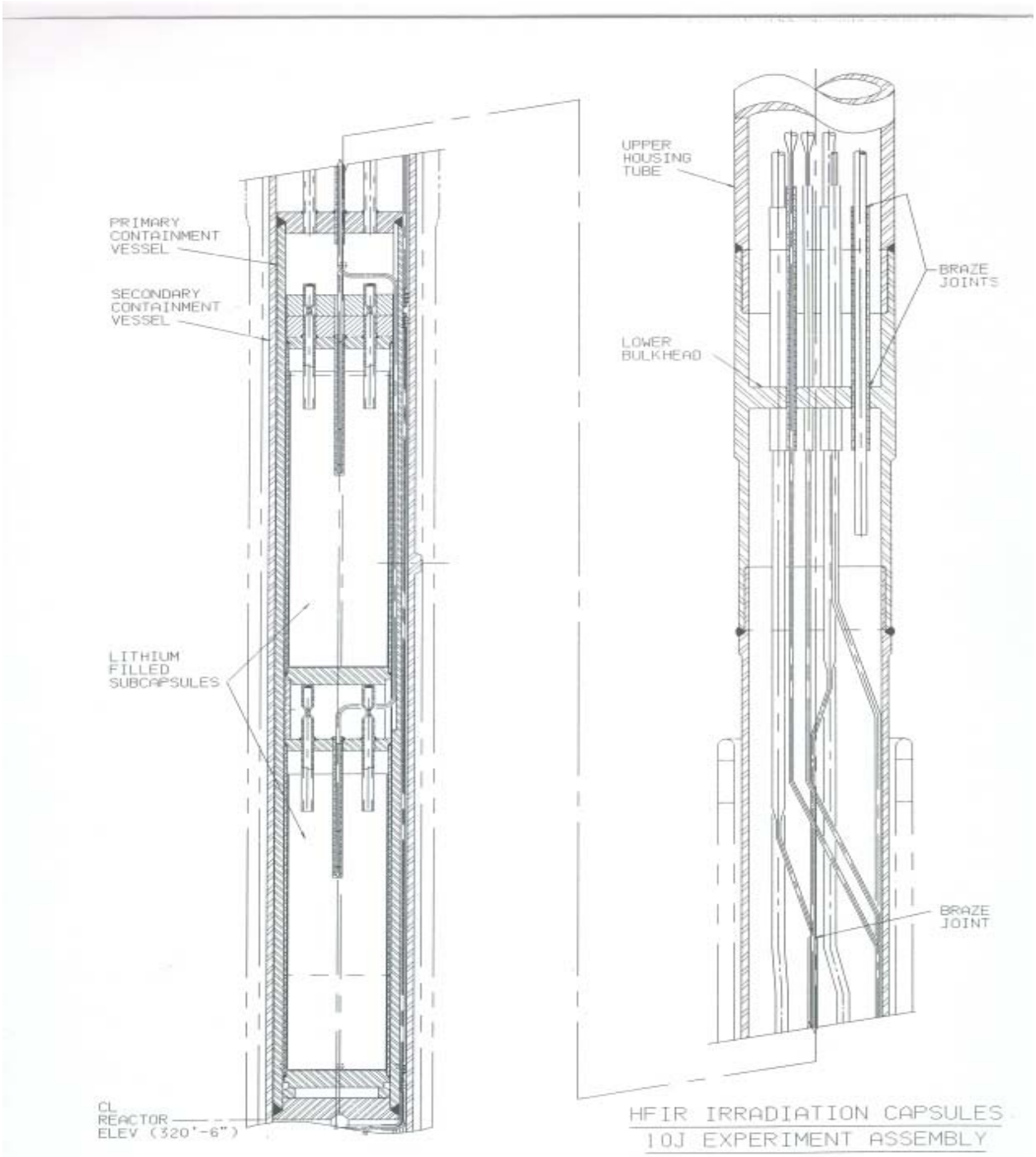


Figure 2. Arrangement of RB-10J from core midplane to above lower bulkhead

The initial mode of operation for RB-10J called for the primary containment to operate as a static gas (helium) system at 80 psig, while the secondary system was a continuously flowing mixed gas (helium and neon) operating at 30 psig. Irradiation began on October 13, 1998, and after about 10 days of normal operation the pressure in the primary system began to decrease indicating that a leak had developed somewhere in the primary containment. A series of tests were performed to try to determine the location of the leak, and at the conclusion of those tests it was apparent that the leak from the primary system was within the secondary containment in the region below the lower bulkhead. The primary suspect for the leak was one of the two brazes that seal the thermocouple assemblies into the primary system. Subsequent examination of these braze joints in the hot cell, discussed in detail in Section 3.1, confirmed that one had developed a leak.

In accordance with the experiment approval documentation, the leak required that the reactor be shut down and the experiment removed. This was done on October 23, 1998; the experiment was placed in the in-pool experiment storage facility while remaining connected to the instrumentation facility.

After modifying the mode of operation and performing additional safety analyses RRD and the RERC approved reinstalling the experiment in the reactor. Irradiation resumed on August 11, 1999. The mode of operation was changed to have the primary system operating statically at 470 psig and the secondary system operating as a mixed gas swept system at 490 psig. The existing very small leak between the primary and secondary systems now allowed small amounts of gas to leak from the secondary into the primary, and the difference between the inlet and outlet flow rates of the secondary system would serve as an indication of either an increase in the existing leak, or a leak in the secondary system. A total of about 150 standard cubic centimeters per minute (sccm) was being flowed through the secondary system and because of system dynamics the difference (outlet flow minus inlet flow) varied between +5 and -10 sccm. This pattern continued through the first nine days of operation, then began to decrease, reaching about -30 sccm after 15 days of operation. This was reviewed by RRD and the RERC and while both approved continued operation with some additional monitoring, an administrative decision was made to shut down the reactor and remove the experiment on August 27, 1999. After removal the experiment remained connected to the instrumentation facility until October 6, 2000. It was then disconnected from the instrumentation facility and moved to the spent fuel pool where it remained until June 5, 2001, when it was shipped to the hot cell.

2.2 RB-13J

The RB-13J experiment was an extremely complex experiment consisting of four specimen holder regions. Through the use of mixed gases (helium and neon) and electrical heaters the temperature of each region could be controlled independently. Five 1/16 in. gas lines supplied the temperature control gases and helium purge gas to the experiment. Tubing of sufficient length was not readily available at the time of assembly of the experiment and therefore couplings were used to join two lengths of 1/16-in. tubing for each of the five gas lines. The tubing was joined to the couplings by brazing in accordance with ORNL brazing procedure BPS-401.

The irradiation of RB-13J began July 22, 1998, and the performance proved to be outstanding for eight cycles. After the eighth cycle, during the EOC 369 refueling outage on May 17, 1999, the capsule was removed from the reactor and placed in the storage facility, as is done every refueling outage. On May 25, 1999, at approximately 11 p.m., the capsule was installed into the reactor in preparation for the ninth of ten planned cycles of operation. The following morning the capsule pressure was 0 psi, and there was no flow indicated through the capsule effluent line. This implied that a leak had occurred in the capsule.

After performing several tests and attempting to control the flows and pressure in the experiment using various techniques, it was surmised that there was a good possibility if irradiation was resumed, it would not be possible to control the temperatures as well has they had been up to that point. Because the customer preferred to accept the shortened irradiation time over the risk of less precise temperature control the irradiation of the experiment was terminated.

2.3 In-pool and dry-wall junction boxes

As stated in the above section, when it became impossible to maintain appropriate flow and pressure conditions in RB-13J, several alternative modes of operation were investigated. One of the options considered involved controlling the atmosphere inside the in-pool and dry-wall junction boxes. The historic method of operation of these facilities is to maintain an ordinary air atmosphere vented to the HFIR open hot off-gas (OHOG) system. It was thought that by maintaining a helium atmosphere at a slightly positive pressure, it might be possible to control temperatures in RB-13J well enough to continue irradiation. To assure that the proposed helium atmosphere would be

reasonably pure, a series of evacuation and back fill operations were performed to remove as much air as possible. At the time these operations were performed, there were four experiments connected to the in-pool junction box; RB-10J, RB-13J, RB14J, and TRIST-TC1.

The process of evacuating the boxes by pulling a partial vacuum apparently opened a leak in the in-pool junction box, which resulted in partially filling both boxes with pool water. The leak probably developed in one of two gaskets used in the in-pool junction box. While the boxes are designed to handle a small pool-water leak by venting it to the process liquid drain line, the valve to that line was closed to facilitate the evacuation and back fill process. Under normal conditions, water would be drained to the process waste line before it could reach a level that would allow it to enter the umbilical hoses that run between the in-pool junction box and the top of each experiment. However, because the valve to the process waste line was closed, the water level got high enough to fill the umbilical of the TRIST-TC1 experiment. While water was not visible in the umbilical hoses of the other experiments attached to the box, it was impossible to know for sure whether or not water had passed through the hoses and was in the solid pipe region of the experiments above the lower bulkhead.

Through a process of manipulating the TRIST-TC1 experiment and its umbilical hose, most of the water was removed and drained through the valve boxes. It was felt that while it was not desirable to have water in the region above the lower bulkheads of the experiments, the stainless steel components in this region would not be in jeopardy, and eventually all of the water would evaporate into the OHOG system.

After the decision was reached to not continue the operation of RB-13J, the operation of the junction boxes was returned to what it had been from their inception, i.e., simply vented to the OHOG.

2.4 RB-14J

The irradiation of RB-14J was begun on June 3, 1999. This was also a very complex experiment with four independently controlled regions using mixed gases (helium/neon or helium/argon) to control temperatures. The experiment operated as it was designed to, but because of the problems experienced with RB-10J, RB-13J, and some rabbit capsules, an administrative decision was made to temporarily terminate the irradiation on August 27, 1999, after about three cycles of irradiation. Following several internal reviews and additional safety analyses, approval was granted to reinstall the experiment, and irradiation was resumed on January 27, 2000.

After an additional three cycles of operation, erratic moisture indications began to be registered by the two moisture monitors on the capsule effluent gas system. These continued during the subsequent cycle, however, because it could not be determined for certain what the cause of the erratic moisture indications was, and because small amounts of mercury were found on one of the moisture probes, it was decided to permanently terminate the irradiation after seven of ten planned cycles of irradiation.

3. INVESTIGATION OF OPERATIONAL DIFFICULTIES

3.1 RB-10J

Because the leaks in both the primary and secondary containment systems of the RB-10J experiment were determined to be in the highly radioactive part of the experiment, all of the investigative work had to await moving the experiment into the hot cell. All instrumented experiments are shipped to the disassembly hot cell (building 3525) using the so-called Loop Transport cask, which is both a top- and bottom-loading cask. In preparation for shipment the cask is lowered into the HFIR pool water. The experiment is lowered into the cask through the top opening in such a manner that the top of the experiment is never placed below water. Once the experiment is inside the cask, the cask is raised out of the water and placed on the floor of the HFIR high bay area in a vertical position with the top ~4 ft. of the experiment protruding out the top of the cask. This top section of the experiment is cut off, a top is put on the cask, and the cask is drained and purged with dry air. The cask is then rotated to a horizontal position and mounted onto a truck for transport to building 3525.

The RB-10J experiment was shipped to the hot cell on June 5, 2001. Three days later the first steps were taken to perform a series of tests in the hot cell charging area prior to actually placing the experiment in the hot cell. With the cask still in a horizontal position, the bolts holding the top cover plug were loosened in preparation for removal of the top plug when about 1 liter of water came out of the plug onto the charging area floor. Health Physics personnel checked the water and found it to be contaminated, with the main activity due to Ag-110. The only source of Ag-110 was thought to be the silver brazes in the vicinity of the experiment lower bulkhead, and most likely the Ag-110 was leached into water that had probably been in the upper housing tube since water was allowed to enter the in-pool junction box two years prior to this.

Once the area was cleaned up, a series of pressurizations and leak tests were performed to (1) confirm that the leak between the primary and secondary systems was still there, and (2) make certain that there were no leaks in the thermocouple assembly tubes (TCATs). After both of these were confirmed the capsule was placed in the hot cell. The first braze joints to be examined were those just above the lower bulkhead. This was done by

cutting through the upper housing tube about an inch above the lower bulkhead and pulling the upper housing tube up far enough to see the tube-to-tube brazes and the bulkhead brazes. Then argon gas was used to pressurize the secondary containment vessel, but the leaks in these brazes were so large that only 40 psig was used, along with a soap check solution. Large leaks were observed in two of the tube-to-tube brazes above the lower bulkhead, and a videotape documentation of these leaks was made. These leaks were obviously much larger than they had been when the first indications of leaks in this system were observed in August of 1999.

To observe the two braze joints of the primary containment system, the secondary containment vessel was cut below the lower bulkhead and removed from the primary containment vessel. The entire primary containment vessel was leak tested by pressurizing the inside of the vessel to 100 psig and putting a soap solution over the entire vessel including all weld joints and the two braze joints. The only indication of a leak was at one of the braze joints, which showed a very small amount of foam after a few minutes, at which time the soap solution usually dried. As a more definitive test, the entire primary containment, still pressurized to 100 psig with argon, was submerged in water up to the cut below the lower bulkhead. In this test the only observed leak was a very small steady stream of bubbles coming from the braze joint that was determined to leak earlier with the soap solution method. This leak test was also recorded on videotape and is available for viewing.

To further determine the cause of the leaks in the braze joints both above the lower bulkhead and in the primary containment system, four braze joints were removed to make metallographic mounts. The four braze joints chosen were the two (one with a leak and one without) in the primary containment system, and two that leaked above the lower bulkhead. All four of these were tube-to-tube joints (or thermocouple-to-tube joints which are similar), and Figure 3 is a sketch showing the joint geometry and the features of an acceptable braze joint.

As can be seen in Figure 3, the braze material should wet the annulus between the two tubes and there should be a fillet where the inner tube enters the outer tube. To confirm the presence of good wetting of the annulus, the metallographic mounts of the braze joints where made by grinding the joint from the top to the point where the fillet was ground

away such that we could see the very top of the tube-to-tube annulus. The results of this process for the primary containment brazes is shown in Figure 4. The leaking joint is shown in plate 6179-07 (or LT), and the non-leaking joint is 6178-07 (or ST). It is quite obvious from Figure 4 that there was insufficient wetting of the annulus in both the leaking and non-leaking joint. For the non-leaking joint therefore, the fillet was apparently all that prevented that joint from also leaking. Similar metallographic mounts (shown in Figure 5) were made of the tube-to-tube braze joints above the lower bulkhead, and they show the same result.

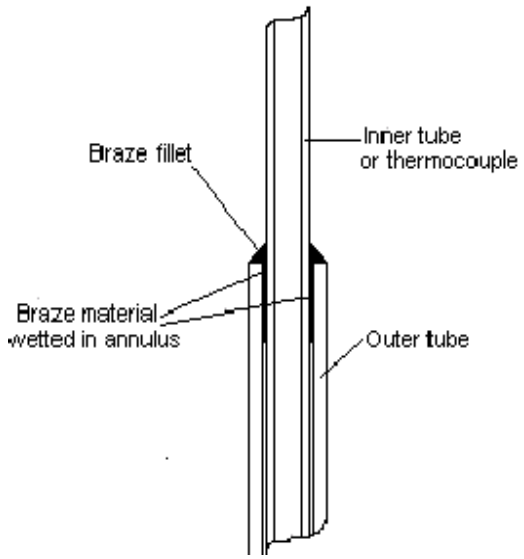
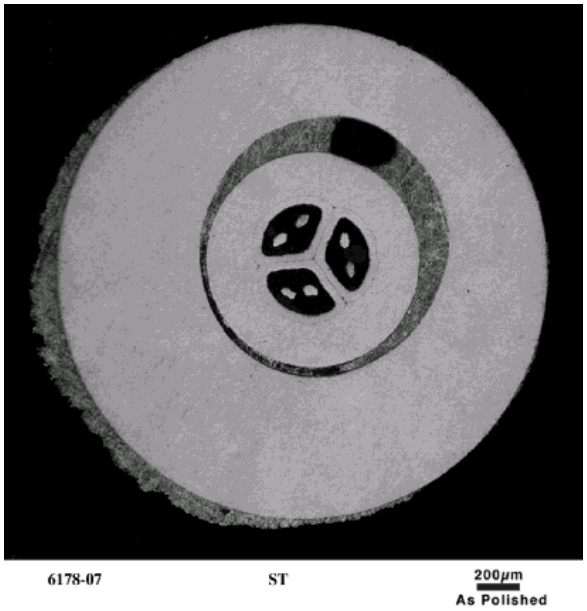


Figure 3. Typical geometry of an acceptable tube-to-tube braze joint



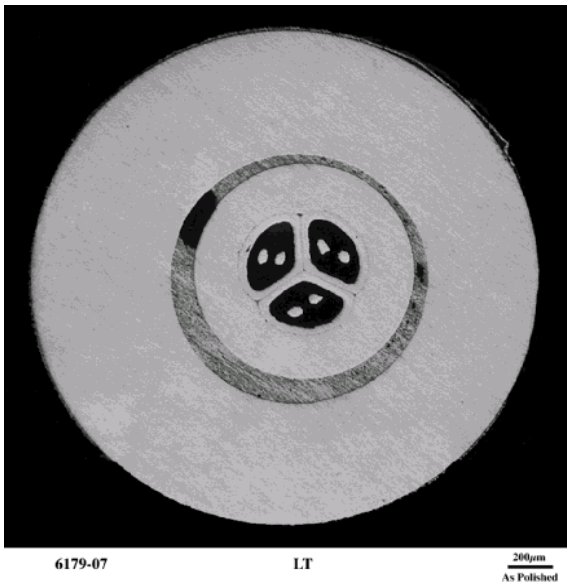


Figure 4. Micrographs of the two braze joints in the RB-10J primary containment system showing lack of penetration of braze material in annulus.

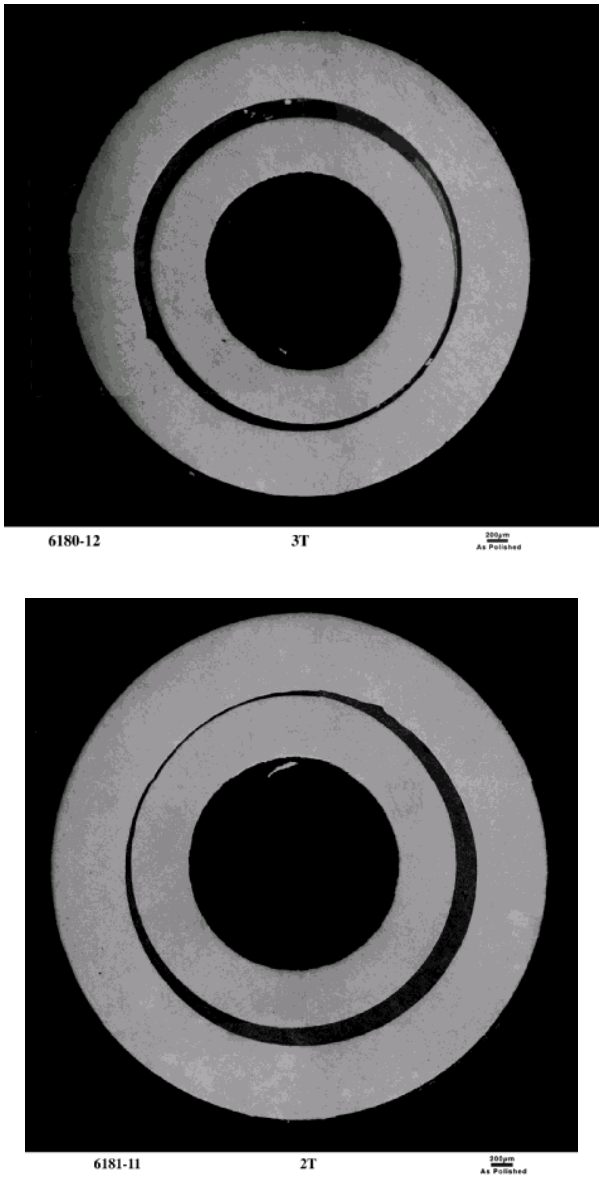


Figure 5. Micrographs of two braze joints just above the lower bulkhead in RB-10J also showing lack of penetration in the annulus

3.2 RB-13J

Tests performed on the RB-13J experiment when pressure could not be maintained in the capsule seemed to indicate that a leak had developed in one of the 1/16-in. gas supply lines. A primary suspect for the location of a leak was the brazed coupling-joint used to join two lengths of tubing for each of these lines. After the experiment was separated from the instrumentation facility and the umbilical hose, these joints were recovered, and

in one of them the tubing had pulled completely free of the coupling into which it had been brazed. The tube still had the braze fillet on it, but there was no braze material below the fillet, in the annulus between the tube and the coupling as there should have been. It was postulated that the manipulation of the experiment to return it to the reactor in preparation for another irradiation cycle put a strain on what was a poor joint and the tube was dislodged from the coupling. Experiments assembled after RB-13J used continuous lengths of 1/16-in. tubing and therefore do not have this braze joint.

3.3 In-pool and dry-wall junction boxes

The problem with getting water into the junction boxes was attributed to the pulling of a vacuum on the system, and probably dislodging either the flat gasket on the in-pool junction box cover, or the gasket between the junction box and the pool wall penetration flange. (It is also possible that the seals used on the bushings leading into the junction box developed leaks.) After the water was drained from the boxes they were pressurized to about 10 psig for several hours, then returned to their normal mode of operation, i.e., vented to the OHOG. A liquid level detector was added to the instrumentation monitoring the boxes, and no additional liquid has been detected to date.

3.4 RB-14J

The RB-14J experiment was sent to the hot cell about 5 months after irradiation was terminated. Prior to disassembly for specimen retrieval the upper housing tube was cut just above the lower bulkhead to facilitate examination of the braze joints in that region. While the braze joints themselves appeared to be satisfactory, there was an appearance of corrosion products on the tubes, with the concentration of these products seeming to be at the same elevation. This observation leads the author to postulate that water was present in the region just above the bulkhead and the corrosion products were deposited on the tubes from the level of the water upward. It is most likely then, that the erratic moisture monitor indications were a result of water above the lower bulkhead and poor braze joints. A photograph showing the appearance of the tubes in this region can be seen in Figure 6.

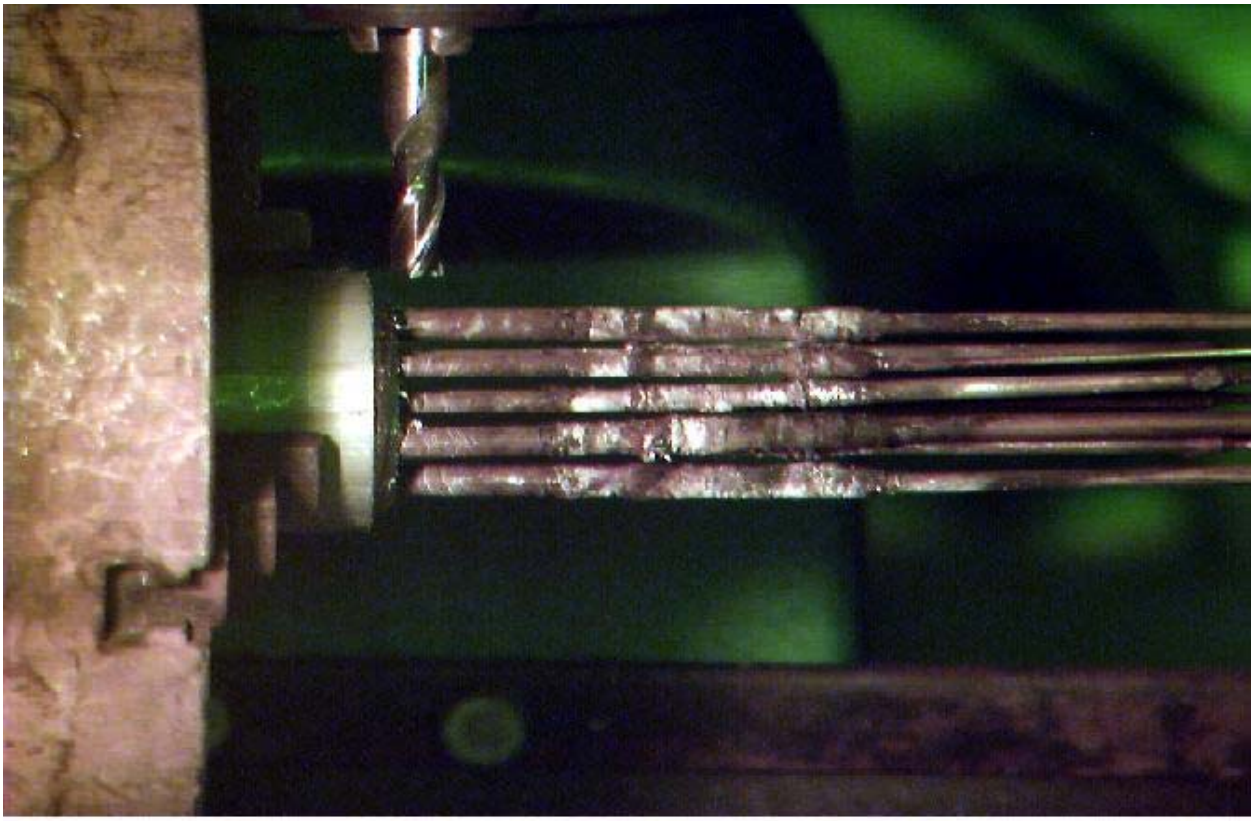


Figure 6. Photo of tubes in region above lower bulkhead in capsule RB-14J

4. CHANGES BEING IMPLEMENTED TO PREVENT RECURRENCE OF OPERATIONAL DIFFICULTIES

After reviewing all of the operational difficulties and investigations into causes of the problems, it appears that the problems can be attributed to poor braze joints, and having water in the region just above the lower bulkhead. To eliminate recurrence of these problems several changes will be incorporated in future irradiation experiments. The first involves a redesign of braze joints used in tube-to-tube brazes. Development of this new joint design is discussed in Section 4.1. Additional changes to be incorporated include relocating the lower bulkhead and changing the atmospheric environment in the region of the capsule between the lower bulkhead and the top of the reactor vessel head.

4.1 Braze joint design

As stated in Section 3.1, metallographic mounts of braze joints have shown that there has been insufficient wetting of braze material in the annulus of tube-to-tube joints. These brazes have passed inspections by passing a helium leak test, and a visual inspection in which the inspector examines the fillet at the top of the joint. However, inspection techniques have not been able to show the extent of braze metal penetration into, and wetting of, the annulus. The tubes and TCATs in these joints are fixed and therefore it is not possible to apply brazing flux in the annulus where it is needed to assure that a good joint can be completed. To avoid this problem a new joint design was necessary that (1) would not rely on being able get flux into the annulus between the two fixed tubes, and (2) could be inspected to assure that a good braze joint had been completed.

A proposed solution that was adapted was to place a sleeve over the original joint. It was decided to test both a tapered brazing sleeve, which would allow matching material wall thicknesses, and a straight brazing sleeve, which would allow heating the joint more uniformly. Figure 7 is a sketch of how such joints would appear in longitudinal cross section.

This design no longer relies on braze material wetting the annulus between the two tubes that are being joined. To demonstrate this technique, several sleeves 3/8 inches long were fabricated from stainless steel rod. The sleeves were drilled 3/16 inches into either end using drill sizes that were a few mils larger that the tubes in the joint. Flux was applied to the entire joint area and green stop-off was applied to the outside of the sleeve to prevent the braze metal from wetting around the sleeve. The sleeve was then slid down over the joint and onto the brazing flux and brazing flux was applied thoroughly to both ends of the sleeve. The braze was made by applying braze material to one end of the sleeve only, and letting the heat from the torch force the braze material to wet thru the sleeve and out the other end. The appearance of a fillet of braze material on the opposite end would be evidence of complete wetting of the annulus under the sleeve. Wetting of the annulus between the original tubes, where brazing flux could not be applied, would not be necessary. Following, in Figure 8, are pictures of braze joints that were completed in the manner described.

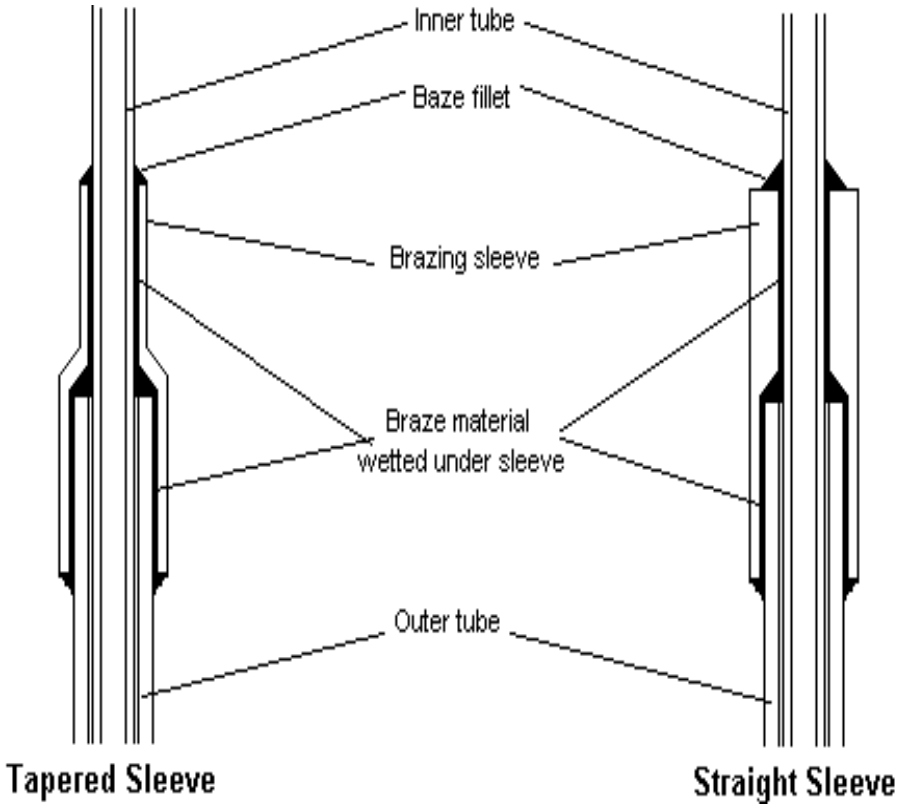


Figure 7. New joint designs for tube-to-tube brazes

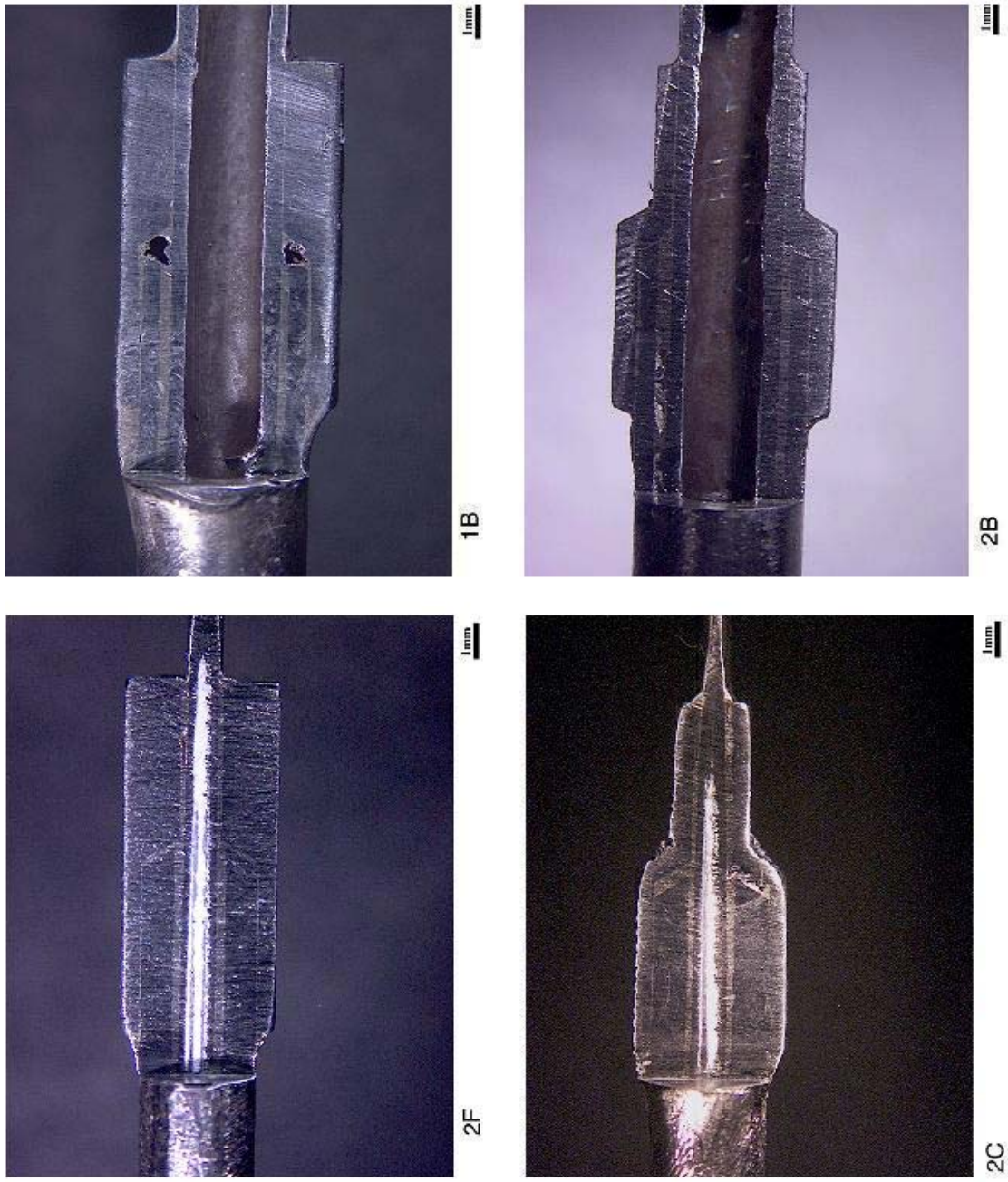


Figure 8. Photos through cross sections of test braze joints. Clockwise from upper left (1B) tube-to-tube with straight sleeve, (2B) tube-to-tube with tapered sleeve, (2C) thermocouple-to-tube with tapered sleeve, and (2F) thermocouple-to-tube with straight sleeve.

Two additional test brazes are shown in Figure 9 with parts of the joint identified.

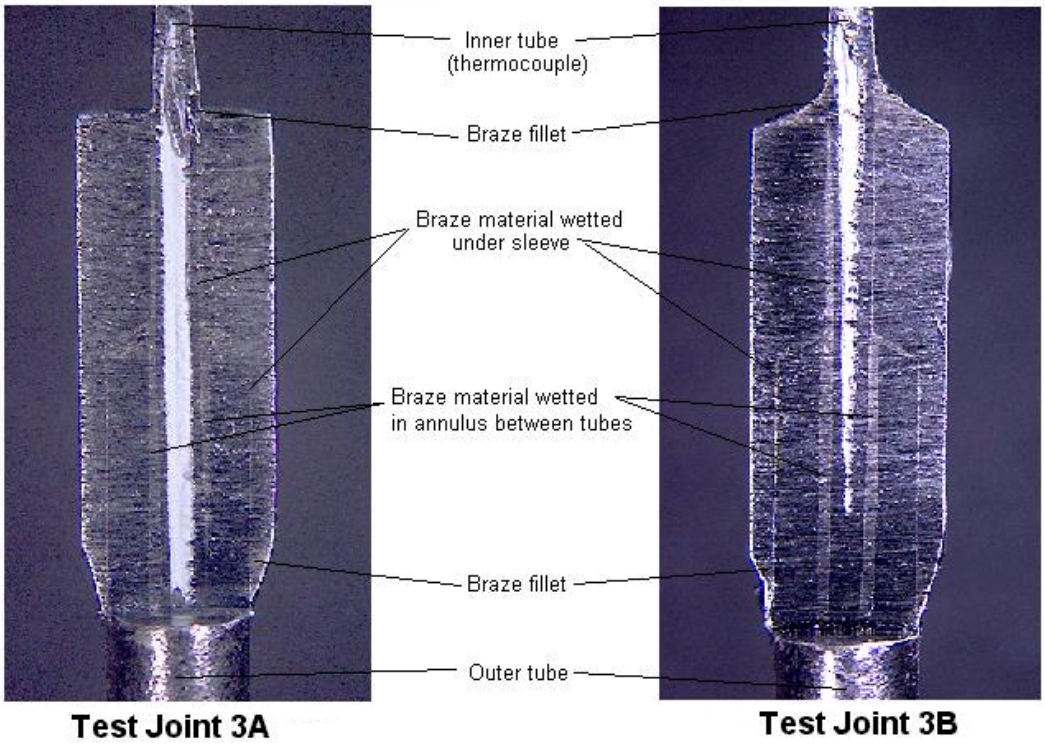


Figure 9. Two thermocouple-to-tube test brazes with straight sleeves

The photographs show that the brazing material not only wetted the annulus between the external sleeve and the tubes, but it also wetted the annulus between the tubes where the braze material was originally desired. A total of 12 such joints were prepared. After successfully completing the first six joints to prove wetting could be accomplished, 6 additional joints were completed, steamed cleaned, successfully helium leak tested, and sectioned to examine wetting of the joint.

It was concluded in these tests that application of brazing flux into the annulus between the tubes in the joint was not required in order to wet the annulus with braze material to produce an acceptable joint. It appears that the brazing flux and braze material will enter and wet the annulus for quite a depth if the joint is slowly and properly heated to drive the materials to the desired location. Unfortunately, there are no non-destructive methods to determine the depth of wetting of such braze joints. This makes the addition of the

external sleeve even more important. It was determined that with the additional sleeve and application of braze material to only one end, an acceptable joint would be obtained with wetting throughout the sleeve, as evidenced by the fillet of braze material on the end of the sleeve opposite where the material was added. Also, as an unexpected benefit, heating the joint properly to get the braze material to flow from one end of the external sleeve to the other forced the brazing flux and material to wet into the annulus where the braze material was originally desired. In other words, addition of the sleeve on the outside serves dual purposes. When wetted with braze material from one end of the sleeve to the other an acceptable braze seal can be obtained. Also if the joint is properly heated to force the braze material to wet under the entire sleeve, the braze material also will enter and wet the annulus where the braze seal was desired originally. The brazed sleeve on the outside is external evidence of what happens on the inside.

Based on this braze development project the following changes will be incorporated into the brazing procedure to be used for all braze joints in future materials irradiation experiments.

1. External sleeves (~ 3/8 in. long) will be used on all seals of tube-to-tube or thermocouple-to-tube joints.

2. Green stop-off will be painted on the outer surface of the sleeves to prevent braze material from reaching the other end by wetting the outer surface.

3. Brazing flux will be applied to the joint before sliding the sleeve into place, and once in place, flux will be applied to both ends of the sleeve.

4. The braze joint is to be heated thoroughly and slowly, allowing the brazing flux ample time to wet and prepare the metal surfaces before the braze material is applied.

5. The braze material is to be added only to one end of the sleeve and heat should be used to force the braze material thru the sleeve and out the other end forming a smooth fillet at both ends of the sleeve.

6. The completed braze joint will be cleaned with steam while under a pressure differential of about 15 psig to remove any residual flux from the joint.

7. The joint will be helium leak tested after it is steamed cleaned. If a leak is discovered the joint should be covered with brazing flux, heated to the remelt temperature, and additional braze material added if necessary. The repaired joint should be cleaned again using steam and pressure before the helium leak test is repeated.

4.2 Capsule design

While there was no evidence that either neutron damage or elevated temperature contributed to the braze failures, steps will be taken to reduce the possibility even further. The lower bulkhead will be moved up approximately 10 inches from where it was in these capsules, returning it to the location it was for other experiments that were run successfully for up to 22 HFIR cycles. This will significantly reduce the neutron flux and gamma heating rate at the location of the lower bulkhead and the brazes immediately above it. Also, this will allow any brazes below the lower bulkhead (e.g., the primary containment brazes in RB-10J), to be located in a region with lower neutron flux and gamma heating. They will now be made as high as possible, and steps will be taken to minimize the temperature that these brazes are subjected to during normal operation.

Additional gas lines (and still-to-be-determined instrumentation) will be added to the region between the lower bulkhead and the in-pool junction box to provide a means to (1) sweep the region with an inert gas, (2) determine if there is a leak in either the lower bulkhead or the tube-to-tube braze joints above the lower bulkhead, (3) determine if there is water on top of the lower bulkhead, and (4) remove any water that might accumulate on the lower bulkhead.

4.3 In-pool and dry-wall junction boxes

During the beryllium changeout outage the two gaskets used in the in-pool junction box were replaced. One of these gaskets was originally installed when the facility was built about 30 years ago. While the operation of these junction boxes will continue as before, the atmosphere in the solid lead tube portion of experiments that vents to the boxes, i.e., the region between the lower bulkhead and the top of the reactor vessel, will be controlled through a still-to-be-determined method. The liquid level detector installed after water had partially filled the boxes in May 1999 will become a permanent part of the junction box monitoring system. Other seals used in the in-pool junction box (e.g., O-ring seals on bushings and gasket seals on caps) will be closely examined and replaced if necessary.