# Development of a Rebrazeable Containment System for Special Nuclear Material Storage and Transport*

*J.D. Pierce, J.J. Stephens, C.A. Walker, F.M. Hosking, R.M. Curlee*
*Sandia National Laboratories***

## INTRODUCTION

A novel means of closing and sealing small type B radioactive material transport packages for surface or air transport as governed by 10CFR71 or NUREG-0360 has been developed at Sandia National Laboratories (SNL). This method is a controlled brazing process that may be used to attach and seal a closure lid to a containment vessel and then remove it at a later time. The process may be performed multiple times without the need for special preparations of the braze joint.

A number of advantages for utilization of this technique have been determined. A brazed seal has integrity at high temperatures for better protection in accident or abnormal environments. A properly designed joint has essentially the same strength as the parent metal. A closure that is brazed, therefore, will no longer be the anticipated point of failure for a broad range of accident environments. This technique will allow the containment vessel design to be optimized with a lighter, more uniform wall thickness throughout. Finally, with a well-defined process for sealing, mechanical inspection, leak testing, and then reopening at a later time, automation of the process is relatively straightforward, and the overall system should be as easy to use as one that utilizes elastomeric seals for containment.

Several technical challenges were addressed throughout the development of this process. The first was to identify a braze alloy that could be used to braze and debraze two metal surfaces repeatedly in an inert atmosphere by only applying new braze material, either as a preformed stamping or a paste. A sound brazed joint had to be made without special pre-braze joint preparation. This made it necessary to identify a brazing process that utilizes inert atmospheres instead of corrosive fluxes to control surface oxidation in order to eliminate the potential for long-term corrosion. A heating process for fast, localized heating that could be accurately controlled to within a few degrees had to be identified and its use demonstrated. In addition, the process could not be cost prohibitive and had to utilize non-destructive testing methods for inspection of both the mechanical joint and leak-tightness.

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The cyclic brazing activity has demonstrated the feasibility of brazing and debrazing piece parts together in a helium atmosphere for as many as twenty times without the need to perform any special joint preparation. This is an ongoing effort with multiple phases that to date has focused on metallurgical and processing issues. Current and future activities are directed toward design issues and an applied demonstration. The first phase of the study required an initial selection of braze alloys with melt temperatures in the range of 900 to 1,100 °C and inert atmospheres that could be used with stainless steel materials. The next phase of the study evaluated materials compatibility and braze alloy/substrate reactions when joining two pieces of 304 stainless steel together and demonstrated methods for stabilizing the metallurgy of a leak-tight joint throughout multiple brazes. An optimized cross section of a brazed joint for a container was developed in the third phase. The final phase will demonstrate the use of cyclic brazing on a full-scale prototype container. This will include the development of a demonstration unit using a high-frequency induction heater, high-vacuum equipment, and a helium leak detector.

## BRAZE ALLOY/INERT GAS STUDY

The first phase of the study required an initial selection of braze alloys and cover gasses that could be used with stainless steel materials. The alloys that were selected for the initial wetting studies were 50Au-50Cu, 35Au-65Cu, 82Au-18Ni, and 80Au-20Cu. The wetting studies were performed in a brazing furnace with no flux material present, with atmospheres of hydrogen, argon, helium, a 10% argon/hydrogen mixture, and a 10% helium/hydrogen mixture. Coupons of Nickel-200 ("Ni-200") were used as the parent material for this portion of the study. Ni-200 was chosen rather than 304SS because of the latter alloy's tendency to form a chromia scale in a helium atmosphere at elevated temperatures. The coupons were evaluated for how the braze alloy "flowed" and the amount of discoloration observed on the coupon and the braze material. The gas mixtures which contained hydrogen actually provided the best wetting results. However, since hydrogen gas generation in shipments of nuclear materials with organic residues present has been a contentious issue in the past, it was decided to not introduce any level of hydrogen into the containment vessel if possible. Very acceptable wetting results were obtained with the high-purity helium gas if the chamber was purged adequately to eliminate any presence of oxygen. It was concluded that the all of the braze alloys being evaluated could be brazed to Ni-200 using a pure helium atmosphere. This is also beneficial since helium will be present in the containment vessel and a helium leak detector may be used to quantify the hermetic seal.

## PRELIMINARY JOINT EVALUATION

Preliminary metallographic analysis determined that base-metal dissolution and grain growth are significant problems when repeatedly brazing two pieces of 304 stainless steel together. This undoubtedly caused the material properties of the 304SS to change in the braze-affected region. In addition, the braze alloy would change in composition due to dissolution of elements from the stainless steel, therefore the liquidus temperature would also change. A stable interface which would prevent dissolution of the 304SS and would maintain its physical properties after repeated exposure to the braze alloy at its melt temperature was required. Nickel was predicted to be a reasonable material for this stable interface. This concept was evaluated by first using a braze-deposited surface of BNi-3 alloy (nominal composition in wt.%: 92.4 Ni-3.1B-4.5 Si) to provide a nickel-rich boundary layer on the 304SS. Metallographic analysis of repeated braze and debraze cycles indicated that the nickel zone provided a significant improvement; however, a thicker layer of nickel was needed. These evaluations indicated the braze alloy best suited for rebrazing appeared to be the 82Au-18Ni alloy, which is also identified as the AWS BAu-4 alloy as specified in AWS-A5.8.

To determine the suitability of the BAu-4 alloy for rebrazing nickel parts, a number of piece parts fabricated from Ni-200 were machined and cleaned, and brazing fixtures were fabricated. Multiple pieces were brazed and debrazed 20 times, with a helium leak test performed after each time the pieces were joined. Additional fresh braze alloy was added prior to each rebraze operation; however, no other joint preparation was performed. Sets of brazed parts were destructively analyzed after 5, 10, 15, and 20 cycles. The piece-parts were leaktight ($< 10^{-9}$ std cc/s leak rate of helium) after each braze operation.

## DETAILED BRAZE JOINT DEVELOPMENT

The next task was to study the actual cross section of a conceptual brazed joint for a container. Ni-200 rings were used for the boundary layers in a configuration joining 304 stainless steel test items. A prototype containment vessel was designed using 304SS for the containment vessel and the closure and Ni-200 rings for interface materials. Figure 1 shows a cross section of the prototype container with a brazed closure system. Note that the Ni-200 rings will be brazed to the 304SS parent material using a high-temperature braze alloy. The Ni-200 rings will then be brazed together using a lower melting point braze alloy - one with a maximum brazing process temperature of 1,050°C. While the brazed joint is expected to provide adequate mechanical strength for the closure system, a threaded closure was included as redundancy for this demonstration in anticipation of potential concerns of an extra-regulatory environment. This would provide additional safety by prohibiting the closure from separating from the vessel in the event that the braze alloy would melt in a severe extra-regulatory fire environment.

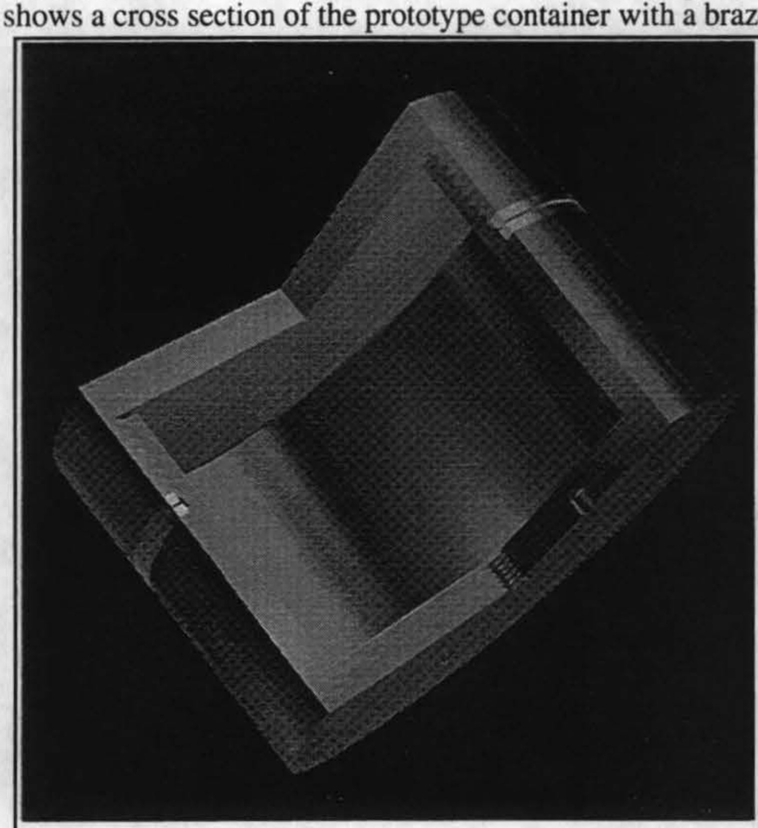

Figure 1. Prototype container with brazed closure. The current container design includes a 304SS vessel and threaded closure, with Ni-200 inserts brazed to both the vessel and closure using the BNi-5 alloy. The two Ni-200 inserts are then brazed/rebrazed with the BAu-4 alloy.

As stated earlier, the cross sections evaluated utilize the BAu-4 (82Au-18Ni) braze alloy in the rebrazeable joint. All Ni-200/Ni-200 braze joints were processed in a He atmosphere, while vacuum or dry hydrogen brazes were used to join the parent metals to the Ni-200 rings.

## Metallurgical Evaluation For 304SS/BNi-3/Ni-200/BAu-4/Ni-200/BNi-3/304SS Configuration

To examine this configuration, small assemblies of 304 SS were fabricated and 1/8-inch thick Ni-200 rings replaced the AWS BNi-3 alloy as the rebrazeable surface. Thus, a step braze configuration was used, where 304SS was first brazed to Ni-200 using the BNi-3 alloy and a braze process temperature of 1,075°C, then the two 304SS/BNi-3/Ni-200 subassemblies were brazed together at the exposed Ni-200 surfaces using BAu-4 alloy. A number of assemblies were fabricated in this manner, and an analytical examination was performed on cross sectioned samples taken from an as-brazed assembly, and assemblies debrazed/rebrazed 5, 10, and 20 times, respectively.

In general, all of the Ni-200/BAu-4/Ni 200 braze joints looked quite sound. Some intermixing of BAu-4 down into the BNi-3 braze joint was observed after several debrazing cycles, although appropriate joint design modifications could undoubtedly avoid this type of problem. Since a new 0.002-inch thick preform of BAu-4 was used during each rebraze cycle, the BAu-4 braze joint increases in thickness after many rebrazes cycles. Figure 2 presents a comparison of brazement thickness data obtained from the metallographic cross sections. The joint thickness increased approximately .001 inch (.0254 mm) for each rebraze.

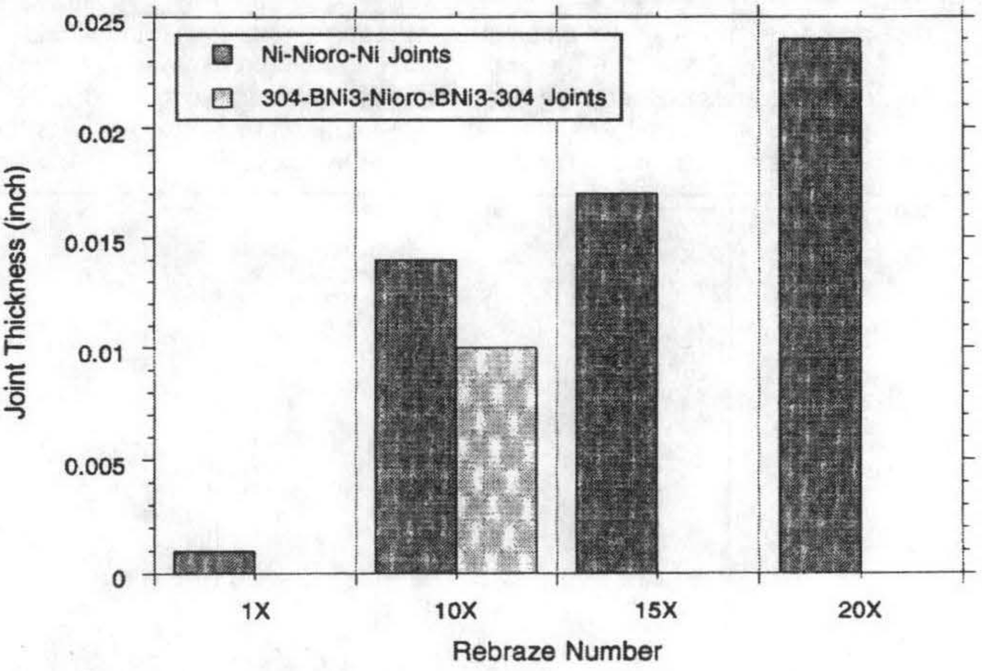

Figure 2. Thickness data for multiple brazes.

The most problematic microstructural feature observed with the 304SS/ BNi-3/Ni-200/BAu-4 configuration is the occurrence of significant amounts of Kirkendall porosity after many debraze/rebraze cycles. Figure 3 shows the line of Kirkendall porosity observed at the 304SS/BNi-3 braze resulting from 20 rebraze/debraze cycles for the BAu-4 alloy, compared with a similar joint in the as-brazed (1 time only) condition.

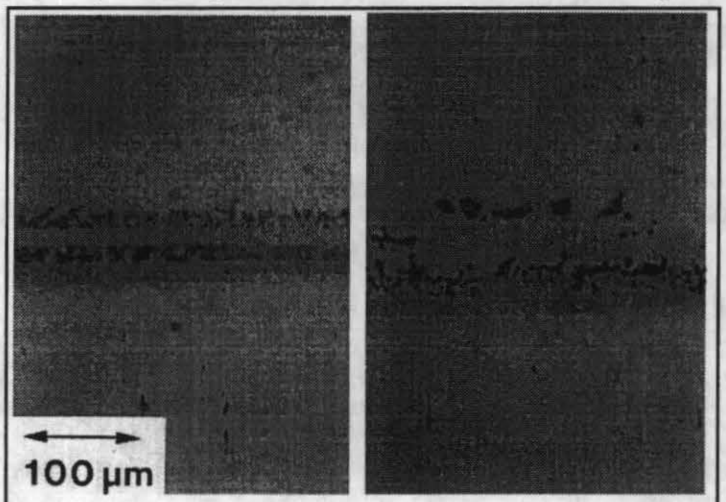

Figure 3. Kirkendall porosity of 304SS/BNi-3/Ni-200 joint following 1 and 20 brazing cycles, respectively.

Electron microprobe linescan results following 20 debraze/rebraze cycles of the Ni-200/BAu4/Ni-200 help to illustrate the advantages of this braze joint configuration. Figure 4 shows an electron microprobe linescan conducted across a portion of the Ni-200/BAu-4/Ni-200 braze joint. While there is undoubtedly some erosion of the Ni-200 substrates, making the joint slightly Ni-rich relative to the original braze preform composition of 82 Au-18 Ni, the only significant feature observed was the increase in braze joint thickness following the 20 debraze/rebraze cycles.

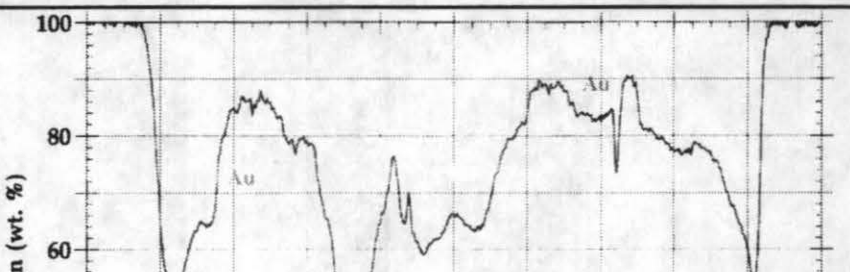

Figure 4. Electron microprobe linescan of Ni-200/BAu-4/Ni-200 braze joint.

## Metallurgical Evaluation For 304SS/"New Braze Alloy"/Ni-200/BAu-4 Configuration

The next task focused on finding a braze alloy for the 304SS/Ni-200 joints which does not possess the Kirkendall porosity problem described above in the 304SS/BNi-3/Ni-200 braze joints. As such, four different "new braze alloys" were evaluated in brazed subassemblies: 65Cu-35Au, BAu-3 alloy (62Cu-35Au-3Ni), BNi-5 alloy (71Ni-19Cr-10Si), and BVAu-8 alloy (92Au-8Pd). All of the individual samples were brazed at their optimum braze process temperatures, then the samples were subjected to 20 separate sequences of simulated braze/debraze thermal cycles which were appropriate for the BAu-4 (82Au-18Ni) braze alloy (peak temperatures for the braze cycle was ~990°C, and the debraze cycle was ~1050°C). The sample which evaluated the 304SS/BNi-5/Ni-200 configuration was brazed in vacuum, while the other three alloys were successfully brazed in a dry hydrogen atmosphere.

The experimental results for the BAu-3 braze alloy indicate that relatively small amounts of braze alloy/substrate interdiffusion occur for this materials combination. However, a large volume fraction of porosity was encountered in the BAu-3 braze joints. The uniformly high level of porosity in the BAu-3 joints indicates that, while BAu-3 is a promising alloy for this application, further process work is needed to avoid obtaining porous braze joints with this alloy/materials combination.

The results for 65Cu-35Au indicated that the debraze temperature cycle used for the BAu-4 alloy is apparently high enough to reflow the 65Cu-35Au braze alloy. This was apparent by means of the excessive dissolution of the 304SS surface, with small islands with composition close to 304SS surrounded by the brazement (not shown in this condensed report). These results suggest that the 65Cu-35Au alloy, while inappropriate for the BAu-4 temperature ranges, could be appropriate for a lower temperature debrazeable system. Silver-base braze alloys such as the 72Ag-28Cu alloy ($T_{eu}$ = 780°C) would be an example of such a lower temperature braze alloy.

Results for the 92Au-8Pd (BVAu-8) braze alloy indicated that substantial erosion of the Ni-200 substrate, along with lesser erosion of the 304SS substrate, is observed as a result of the rebraze/debraze cycles for the BAu-4 alloy. Substantial penetration of Au from the brazement into the Ni-200 alloy was observed in the electron microprobe linescans. This suggests that as a result of the debraze cycle for BAu-4, a portion of the BVAu-8 brazement adjacent to the Ni-200 would perhaps liquate. These results suggest that the BVAu-8/Ni-200 system would not be compatible for this particular application.

The BNi-5 braze alloy appeared to be the most attractive of the four candidate braze alloys evaluated. The most significant reaction is the diffusion of Ni from the Ni-200 into the braze joint and diffusion of Cr from the braze joint into the Ni-200. The absence of "problematic" features in the 304SS/BNi-5/Ni-200 configuration following the extensive braze/debraze cycles, along with the good quality of joints (i.e., no large voids observed) suggests that BNi-5 would be the appropriate alloy for this braze joint. A process temperature of 1190° C in vacuum was used for all BNi-5 brazes in this study.

## Metallurgical Evaluation of Nitronic 60/BNi-5/Ni-200 Joint Configuration

If a threaded closure is used with a braze joint, a different material would be desired to prevent galling of the threads on multiple assembly operations. Therefore an investigation was made into using Nitronic-60 material for the closure and 304SS for the vessel body. Detailed evaluations of the Nitronic 60/BNi-5/Ni-200 and the 304SS/BNi-5/Ni-200 braze joints were performed. This was required since coarse particles of Si-containing compounds were observed in the Nitronic-60 braze joint. This was unlike the 304SS/BNi-5/Ni-200 joint which was completely single phase. Extensive characterization using transmission electron microscopy (TEM), electron microprobe, and low-force indentation microhardness techniques ("nanoindentation"), were performed on the Nitronic 60/ BNi-5/Ni-200 braze joint.

All of the samples were assembled using vacuum brazing methods. Following that process, the samples were subjected to 20 debraze/rebraze cycles which were appropriate for the BAu-4 alloy: The high temperature debraze samples were subjected to a debraze cycle with a peak temperature of 1,100° C and a rebraze cycle with a peak temperature of 990°C; the low temperature debraze samples were subjected to a debraze cycle with a peak temperature of 1,050°C and the same rebraze thermal cycle. Figure 5 shows the microstructure of the Nitronic 60/BNi-5/Ni-200 joint, and Figure 6 illustrates the microstructure of the 304SS/BNi-5/Ni-200 joint. Relatively high levels of silicon in the Nitronic 60 (4.5 wt.% maximum) vs. 1 wt.% maximum in the 304SS, appear to be the direct cause of the second-phase particle formation in the Nitronic 60/BNi-5/Ni-200 braze joint.

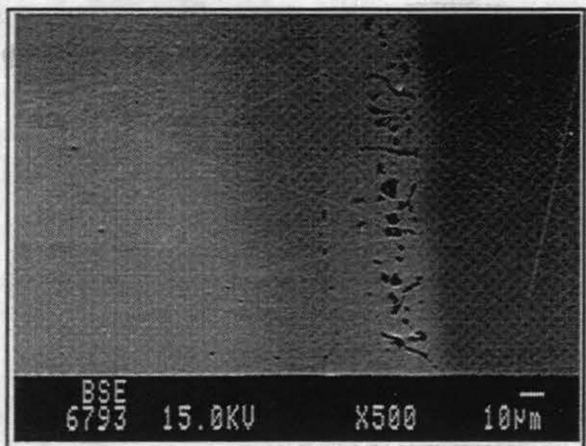

Figure 5. Nitronic 60/BNi-5/Ni-200 joint.

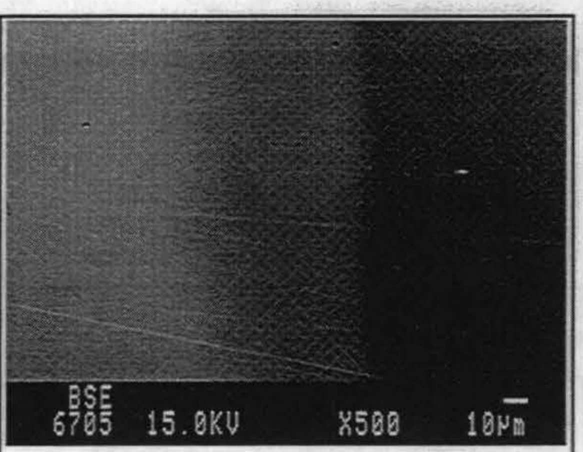

Figure 6. 304SS/BNi-5/Ni-200 joint.

The complex microstructure of the Nitronic 60/BNi-5/Ni-200 joints can be summarized as follows: (a) adjacent to the Nitronic-60 and Ni-200 substrates, coarse, equiaxed particles of $Cr_3Ni_2Si$ are observed; (b) on the Ni-200 side of the joint, a layer of hexagonal $Ni_{31}Si_{12}$ particles is found; (c) immediately adjacent to the $Ni_{31}Si_{12}$ layer, the braze contains fine, ordered precipitates of $Ni_3Si$; (d) except for the occasional coarse $Cr_3Ni_2Si$ particles, the braze is single phase adjacent to the Nitronic-60. Nanoindentation results indicate that the $Cr_3Ni_2Si$ and $Ni_{31}Si_{12}$ phases are quite hard and are expected to fracture in a brittle manner. Because of the potential for brittle fracture in the braze joint, the Nitronic-60 material is ruled out for this braze joint configuration.

Based on all the metallurgical evaluations, the 304SS/BNi-5/Ni-200/BAu-4/Ni-200/BNi-5/304SS material stackup appears to be the best configuration for this application.

## FULL SCALE REBRAZING DEMONSTRATION

A system for brazing, debrazing, and rebrazing full-scale containment vessels for nuclear materials that might be used in a 6M type of shipping container or a storage facility was designed and fabricated. Induction heating equipment is used to provide localized heating for the braze joint. A full-scale vessel was fabricated and sent to a manufacturer of induction heating equipment so that an induction coil could be fabricated and that a power supply could be specified with the proper frequency and power rating. As a result, a one turn induction coil was designed and fabricated and coupled with a 10 kHz power supply capable of providing 50 kW of power. Tests were performed at the manufacturer prior to procuring any equipment to ensure adequate penetration of the brazed area in a short period of time to minimize extraneous heating. Melt temperatures were achieved in 6 to 10 seconds, depending on the power settings used.

A 10 kHz power supply capable of producing 50 kW of power was procured for full-scale brazing experiments to be conducted at Sandia. A brazing chamber was also designed and fabricated that would allow a full-scale vessel to be brazed and debrazed repeatedly in a helium atmosphere and allow helium leak testing to be easily performed on the vessel. Figure 7 shows the equipment assembled in the Transportation Systems Department Laboratory at Sandia National Laboratories.

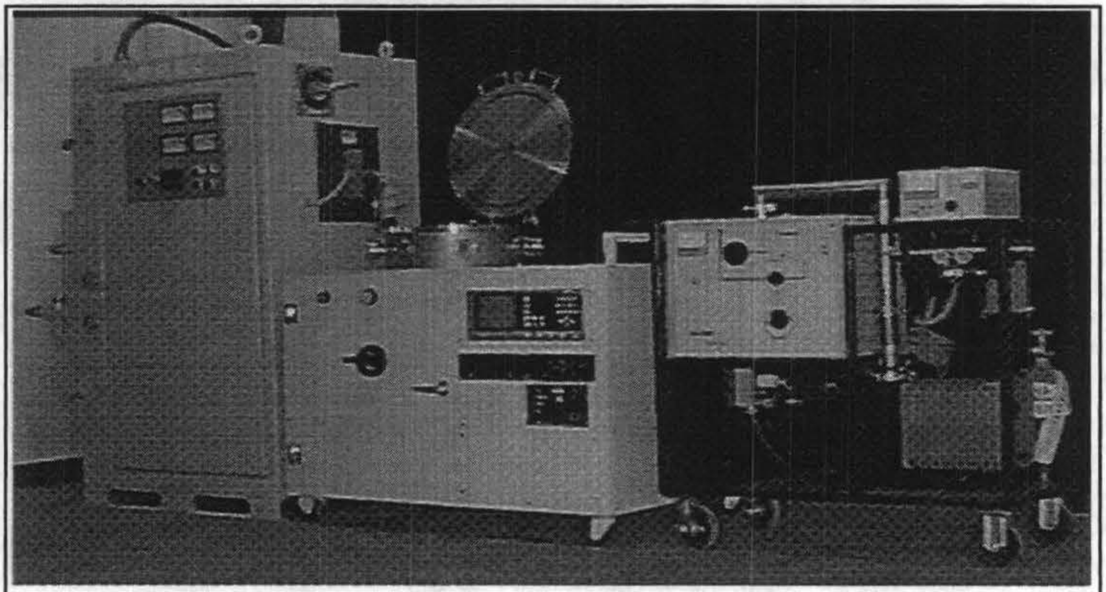

Figure 7. Demonstration induction brazing and leak detection equipment.

Experiments will be conducted in the future using this equipment to evaluate the brazing/rebrazing technique on full-scale containment vessels. In addition, an optimized geometry for brazed joints as well as alternate ways to prevent mechanical separation of the containment vessel and closure in the event of a severe extra-regulatory environment will be developed.

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