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Task 1.5.6 Moderator Containment
Laboratory Experiment Test Plan
(CDRL #5)

for

# SPACE-R THERMIONIC SPACE NUCLEAR POWER SYSTEM

**Design and Technology Demonstration**

Submitted to

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Submitted by

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## Task 1.5.6 MODERATOR CONTAINMENT LABORATORY EXPERIMENT TEST PLAN

### 1.0 OBJECTIVE

The objective of this experiment is to determine the permeation rate of hydrogen from yttrium hydride and zirconium hydride through beryllium in the temperature range of 773 K - 973 K. In addition, Topaz II type zirconium hydride specimens with and without the proprietary oxide coating canned in stainless steel will be tested to measure the hydrogen permeation rate. The TSET SS-canned ZrHx samples currently at Phillips Laboratory will be used for the latter test with Phillips Laboratory participation at the SPI hydrogen leak test stand.

A key technology demonstration of the effectiveness of transferred arc plasma spraying of a 1 mil Molybdenum coating on the Be cladding will be performed. The effectiveness of the Molybdenum coating in preventing any interaction of Be with Stainless Steel in NaK will be assessed and demonstrated.

### 2.0 OVERVIEW

### 2.1 Background

The preferred moderator being considered for SPACE-R is yttrium hydride encased in beryllium tubes. The baseline beryllium performs a dual function as it acts as a moderator and provides containment for hydrogen. The permeation rate of hydrogen from the hydride through the beryllium shell at the operating temperature is an important factor for the functionality and reliability of the Be-YHx moderator. Hydrogen containment capability of beryllium is comparable to enamel which was used in SNAP and Topaz II reactors. See Figure 1. However, limited experimental data base exists for the hydrogen permeation through fabricated beryllium enclosures at high temperature. Permeation of hydrogen in beryllium is strongly affected by surface conditions, thickness of surface oxide, surface and bulk traps, impurity content and microstructure. The permeation rate of hydrogen, measured in recent investigations, in ultrapure beryllium especially after elimination of the effect of surface oxide is 2 orders of magnitude higher than those reported in earlier investigations, as shown in Figure 1.

The beryllium to be used in the SPACE-R moderator is not ultrahigh purity and the permeation of hydrogen is expected to be lower than the data for high purity beryllium of Figure 1. Also, at the maximum operating temperature of 930 K, the dissociation pressure of $YH_{1.75}$ is about .001 to 0.01 torr which is 4 to 5 orders of magnitude lower than $ZrH_{1.85}$. Therefore, the permeation rate of hydrogen from $YH_{1.75}$ clad in Be should be 3 to 6 orders of magnitude smaller than that from $ZrH_{1.85}$ clad in SS and enamel, and about 1 to 2 orders of magnitude lower than the Russian proprietary coating on $ZrH_{1.85}$ and enamel on stainless steel.

The hydrogen containment capability of the $YH_{1.75}$ - Be moderator will be demonstrated experimentally in the prototypic operating temperature range. Test specimens will be fabricated using a procedure similar to that to be used for the moderator of SPACE-R. Zirconium hydride will be tested as an alternate moderator material to be compared with yttrium hydride in Be. Therefore, the permeation of hydrogen in zirconium hydride - beryllium specimens will also be measured for comparison.

The backup moderator for SPACE-R is the Russian $ZrH_{1.85}$ with proprietary coating and a cover gas. As part of the TSET deliverables, Phillips Laboratory currently has Russian zirconium hydride samples. The schematics of these samples are shown in Figure 2. These samples are proposed here to be tested at SPI for the hydrogen permeation measurement to compare the hydrogen containment capability of Be-$YH_{1.75}$ and SS-$ZrH_{1.85}$ at the same temperature and test conditions. The Topaz II hydrogen containment technology is adequate for $H_2$ management at the Topaz II coolant temperature ( max 873 K). However, at the SPACE-R coolant temperature (max 923K), the $H_2$ loss rate from the reactor is estimated to be about 1 % per year with the Topaz II technology. Hydrogen permeation at this higher temperature must be confirmed.

2.2 Test Matrix

A total of 17 specimens will be tested in this task: 5 existing Topaz II specimens and 12 specimens fabricated for this task. Twelve hydride specimens will be fabricated and tested as follows:

Fabricated:

1) Two control specimens : empty beryllium tubes one braze closed and the other EB weld closed for the background measurement.
2) Two $YH_{1.75}$-Be clad specimens with brazed end plugs (Figure 3).
3) Two $YH_{1.75}$-Be clad specimens with EB weld only.
4) Two $YH_{1.75}$-Be clad specimens with brazed end plugs and EB protective closure welds (prototype design).
5) Two $ZrH_{1.85}$-Be clad specimens with brazed end plugs.
6) Two $ZrH_{1.85}$-Be clad specimens with brazed end plugs and EB protective closure weld - reference data.

On $ZrH_{1.85}$ Delivered to TSET by INERTEK/ISP:

A total of 10 samples were delivered to TSET (two samples each of five different configurations). One sample of each configuration will be tested as follows:

1) One type 1 sample in vacuum case without coating, oxidizer or enamel.
2) One type 1 sample in vacuum case with no coating and different solid oxidizer in SS can.
3) One type 1 sample in vacuum case with Topaz II and III protective coating and solid oxidizer in SS can.

4) One type 2 sample with coating, solid $O_2$ source and SS can.
5) One type 2 sample without clad, coating or $O_2$ source.

The hydrogen permeation rate will be experimentally measured for each specimen at 773 K, 823 K, 873 K, 923 K and 973 K.

3.0 TEST SPECIMEN: DESIGN, FABRICATION AND ASSEMBLY

3.1 Preparation of Yttrium Hydride and Zirconium Hydride

3.1.1 The atomic ratio of hydrogen to yttrium of the yttrium hydride to be obtained from Battelle Northwest Lab is 1.9. Zirconium hydride available commercially has a hydrogen to zirconium atomic ratio of about 2.0. These ratios are higher than the 1.75 for the yttrium hydride and 1.85 for the zirconium hydride that will be used in the baseline moderator or the backup moderator of the SPACE-R. Also at these high hydrogen content of the hydrides, the hydrogen pressure will be too high and uncontrollable.

3.1.2 The hydrogen to yttrium ratio will be reduced to 1.75 in the yttrium hydride and the hydrogen to zirconium ratio will be reduced to 1.85 in the zirconium hydride using the procedure previously developed at SPI for this purpose. The hydride will be held in a stainless steel container inside a quartz specimen chamber of the test facility described in section 4.0. The system will be evacuated and the temperature of the specimen chamber will be maintained at a preset value. The butterfly valve will be adjusted to maintain the hydrogen pressure over the hydride at a fixed value to give the desired equilibrium hydrogen content in the hydride. Alternatively, with the hydride inside, the system can be evacuated and then backfilled with Argon to about 1 atmosphere pressure. This pressure will be maintained and the hydride will be heated to a predetermined temperature at which excess hydrogen will be driven off and the desired composition of the hydride will be equilibrium. In either case the hydride will be weighed both before and after the processing to determine the actual hydrogen loss and to confirm the final composition of the hydride.

3.1.3 If necessary, after processing, the hydrides may be analyzed to determine their hydrogen content accurately and independently.

3.2 Specimen Tubes

The Be tube specimens will be prepared by Nuclear Metals from 1 inch OD x 0.050 inch thick wall beryllium tubes as follows:

3.2.1 One 6 inch long control specimen without hydride with brazed caps at both ends and one 8 inch long control specimen without hydride, with electron beam welded caps at both ends.

3.2.2 Two 6 inch long specimens with yttrium hydride and end plugs furnace brazed in helium at both ends.

3.2.3 Two 6 inch long specimens with yttrium hydride and end plugs electron beam welded at both ends.

3.2.4 Two 8 inch long specimens with yttrium hydride and helium backfill, an electron beam welded end plug at one end and a BAg-18 brazed end plug and an electron beam welded end plug closure at the other end.

3.2.5 Two 6 inch long specimens with zirconium hydride and end plugs furnace brazed in helium at both ends.

3.2.6 Two 8 inch long specimens with zirconium hydride and helium backfill, an electron beam welded end plug at one end and a BAg-18 brazed end plug and an electron beam welded end plug at the closure end.

3.3 Be Tube Closure

Counter bores will be machined at the ends of the extruded beryllium tubes, and end plugs will be machined to fit the counter bores. The first end plug will be furnace brazed in 5 specimens and electron beam welded in 7 specimens at one end of the tube. The tubes will be leak checked with a helium leak detector. The tubes will then be filled with hydride which will be held in place by beryllium washers. One control specimen and four specimens of section 3.2.2 and 3.2.5 will be sealed by furnace brazing the second end plug in helium atmosphere. In the four specimens of sections 3.2.4 and 3.2.6, a second end plug will be brazed in place in helium atmosphere using BAg-18 brazing alloy, followed by electron beam welding of a third end plug at the same end. One control specimen and two specimens of section 3.2.3 will be sealed by electron beam welding of a second end plug.

3.4 Molybdenum Coating

3.4.1 One 6 inch long specimen with zirconium hydride and one 6 inch long specimen with yttrium hydride will be coated with 1 mil of molybdenum by Applied Coatings of Columbus, Ohio, using transferred arc plasma spray process.

3.4.2 The effect of transferred arc plasma sprayed moly coating on hydrogen permeation in beryllium will be evaluated and later on, the specimens will be tested for corrosion resistance in liquid NaK in the presence and in contact with stainless steel.

3.5 Due to the low vapor pressure of $YH_{1.75}$ at the operating temperature, a helium bond is required for efficient heat transfer between the hydride and the beryllium shell during normal operation of the SPACE-R reactor. Because electron beam welding should be done in vacuum, the helium bond is achieved in specimens by brazing an extra internal end plug in helium atmosphere. A possibly simpler way of obtaining the helium bond is to laser beam weld the end plugs in helium. Laser beam welding by Lawrence Livermore Lab was given some serious consideration. However, as laser beam welding would require additional development work, it will not be used in the preparation of specimens for this experiment. Note that EB welding or brazing will provide adequate barrier for the dry $H_2$ containment test in this task. Laser beam welding remains as the alternate method of joining the end plugs in the Be moderator assembly of SPACE-R.

## 4.0 TEST FACILITY

The test facility used by SPI in an earlier investigation for hydrogen permeation measurement in zirconium hydride encased in SPI enamel-coated Hastelloy C-276 will be used in this experiment with some modifications. The major difference will be the use of calibrated hydrogen leaks. The schematic of the test facility is given in Figure 4. The specimen will be held in a quartz container which will be placed inside a 3-zone constant temperature electric furnace. The specimen chamber will be evacuated by a turbomolecular pump. Liquid nitrogen cold traps will be used to provide a high quality vacuum by condensing water vapor and other condensable gases.

A quadrupole mass spectrometer residual gas analyzer, a thermocouple vacuum gauge,a convectron vacuum gauge and an ion vacuum gauge will be attached to the system to measure and monitor hydrogen partial pressure and the total pressure in the vacuum system. Two calibrated hydrogen leaks, one in the $10^{-7}$ atm cc/s flow range and the other in the $10^{-8}$ atm cc/s flow range will be attached to the vacuum system just above the specimen chamber. A butterfly valve will be installed in the vacuum line to isolate the specimen chamber and the instrumentation from the pumping system.

## 5.0 INSTRUMENTATION AND CALIBRATION

The Lindberg electric furnace to be used in this investigation is capable of maintaining a constant temperature up to 1200°C within 2°C. It consists of three heating zones with separate temperature controllers to maintain a uniform temperature along the axis with minimum end effects. The temperature of the specimen will be measured by a thermocouple with the hot junction in contact with the specimen.

The residual gas analyzer will be a Transpector model C100M, made by Leybold Inficon. It has both a Faraday cup and an electron multiplier. The minimum partial pressure detectable by this instrument is $5x10^{-13}$ torr. It will come calibrated from the factory. The calibration will be verified by comparing the total pressure read by this instrument with the total pressure measured by the ion gauge. The measurement of the partial pressure of hydrogen will be verified with the calibrated hydrogen leaks.

## 6.0 PROCUREMENT OF MATERIALS AND SERVICES

### 6.1 Hardware for Vacuum System and Furnace

6.1.1 The configuration of the experimental setup has been finalized with regard to the location and type of ports for instrumentation.

6.1.2 New valves, bellows and fittings to be ordered have been identified for the vacuum system.

### 6.2 Calibrated Hydrogen Leak

6.2.1 Two calibrated hydrogen leaks, one in $10^{-7}$ atm.cc/s flow range and the other in $10^{-8}$ atm.cc/s flow range will be provided by Vacuum Technology, Inc.

### 6.3 Residual Gas Analyzer

6.3.1 Leybold Inficon has demonstrated the operation and capability of the Transpector residual gas analyzer attached to a vacuum system. With the calibrated hydrogen leak, the measurement was verified and the upper limit of measurement of hydrogen partial pressure by the Transpector was determined at Leybold Inficon facility in San Jose.

6.3.2 The residual gas analyzer will be leased for the test period for $H_2$ measurements.

### 6.4 Yttrium Hydride and Zirconium hydride

6.4.1 Yttrium hydride will be obtained from Battelle Pacific Northwest Lab and zirconium hydride will be purchased from Teledyne Wah Chang Albany.

## 7.0 TEST PROCEDURE

### 7.1 Set Up and Check Out

7.1.1 The modification and assembly of the test set up will be done as shown in Figure 4.

7.1.2 The system will be pumped down and leak checked with a helium leak detector. The base pressure will be established.

7.1.3 The correlation between the calibrated hydrogen leaks and the hydrogen partial pressure will be determined by measuring the hydrogen partial pressures in the residual gas analyzer in steady state with a calibrated hydrogen leak open to the vacuum system at a time. The rise in the total pressure as measured in the ion gauge and the rise in the hydrogen partial pressure as measured by the residual gas analyzer when the vacuum system is isolated from the pump by closing the butterfly valve will be recorded.

7.2 Product Runs

7.2.1 A control specimen without any hydride will be placed inside the specimen chamber, the system will be pumped down and steady state condition will be established. The total pressure and the hydrogen partial pressure will be recorded. one of the calibrated hydrogen leaks will be opened and the base pressure will be recorded in the steady state. Then the steady state base pressure with the other hydrogen leak will be determined. With the hydrogen leaks closed, the system will be isolated from the pump by closing the butterfly valve and the rise in the total pressure as measured by the ion gauge and the rise in hydrogen partial pressure as measured by the residual gas analyzer will be recorded as a function of time. This will be repeated for the other control specimen.

7.2.2 The specimens with the hydride will be placed in the specimen chamber one at a time and the procedure described in section 7.2.1 will be performed for each specimen.

7.2.3 The experimental processes described in sections 7.2.1 and 7.2.2 will be repeated at temperatures of 773 K, 823 K, 873 K, 923 K and 973 K.

7.3 Data Reduction

7.3.1 The hydrogen permeation rate from the hydride specimens will be calculated from the experimental data using two methods.

7.3.2 The base pressures with the control specimen and opened calibrated hydrogen leaks will be plotted as a function of the value of the calibrated leak. From the value of the base pressure with the hydride containing specimen on this plot, the hydrogen permeation for that specimen will be determined.

7.3.3 The rise in hydrogen partial pressure or total pressure due to hydrogen permeation when the vacuum system is isolated from the pump will be determined by subtracting the rise in pressure with the control specimen from the rise in pressure with the hydride containing specimen.

7.3.4 The hydrogen permeation rate J is given by:

$$J = (22400 / A) * (V / kT) * (dP/dt)$$

where A = Avogadro's Number ($6.022 \times 10^{23}$)
T = temperature in °K
V = volume of the vacuum system in cc
k = Boltzmann's constant
P = pressure
and t = time

7.3.5 The hydrogen permeation rates determined by the above two methods will be compared, analyzed and plotted as a function of temperature for yttrium hydride and zirconium hydride specimens.

8.0 ANTICIPATED RESULTS

8.1 The permeation of hydrogen in the specimens will depend on the test temperature, the hydrogen pressure over the hydride, the purity and surface characteristics of beryllium and the integrity of the end plug joints.

8.2 Based on the available data, the hydrogen permeation rate is expected to be in the range $10^{-7}$ to $10^{-5}$ cc(STP)/hr-cm$^2$ for yttrium hydride specimens and in the range $10^{-5}$ to $10^{-3}$ cc(STP)/hr.cm$^2$ for the zirconium hydride specimens.

8.3 The molybdenum coated specimens may have lower hydrogen permeation rates compared to the uncoated specimens. Molybdenum coating is used not for $H_2$ containment, but for compatibility with NaK and stainless steel when the moderator is used in the reactor. This compatibility will be demonstrated in the program.

9.0 TEST SCHEDULE AND MILESTONES

9.1 The test schedule and milestones are shown in Figure 5 based on the anticipated EAR approval by January 1, 1994. Figure 6 shows the cost breakdown.

10.0 TEST MANAGEMENT AND MONITORING

10.1 Dr. Gauri Das will be responsible for conducting this experiment. He will be assisted by Dave Troetschler. The task progress will be monitored internally by Dr. Kent Koester.

10.2 Monthly reporting will be provided as part of program Monthly Reports. A Final Report will be delivered within 30 days of the test completion.

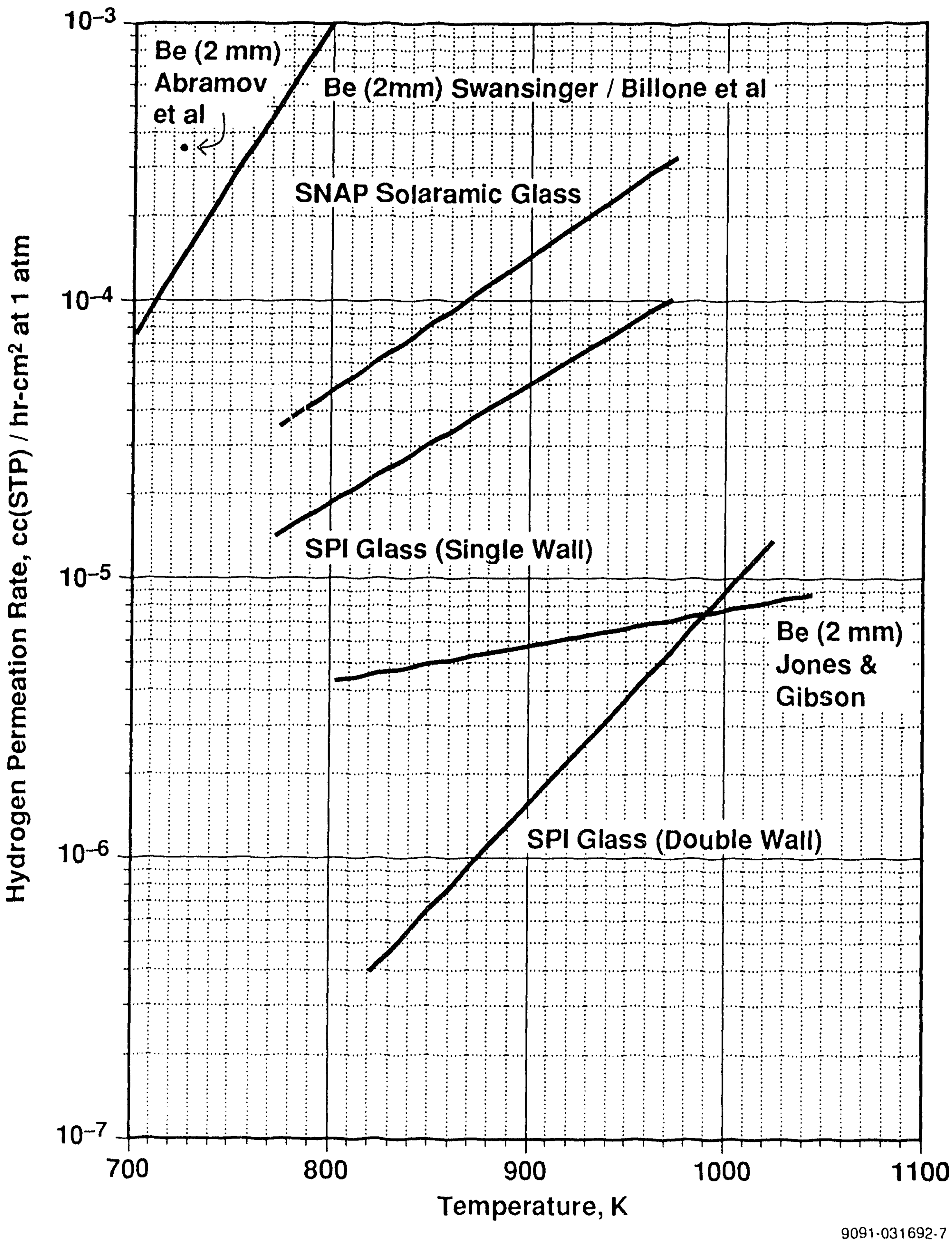
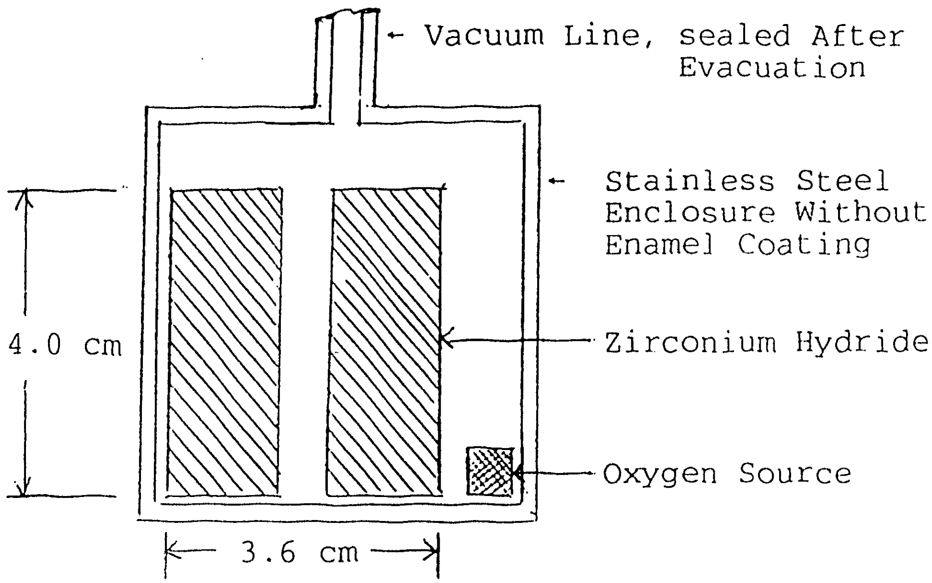
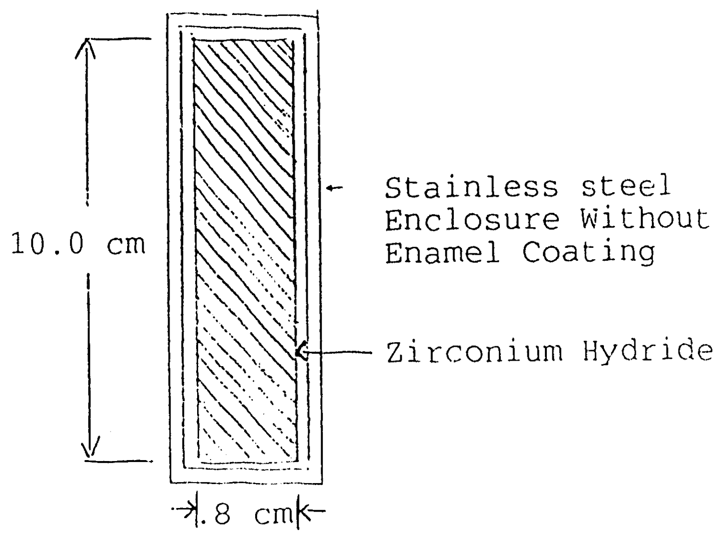



Figure 1. Hydrogen Permeation Through SPI Glass




Type 1




Type 2

FIGURE 2. Schematics of Russian Zirconium Hydride Samples

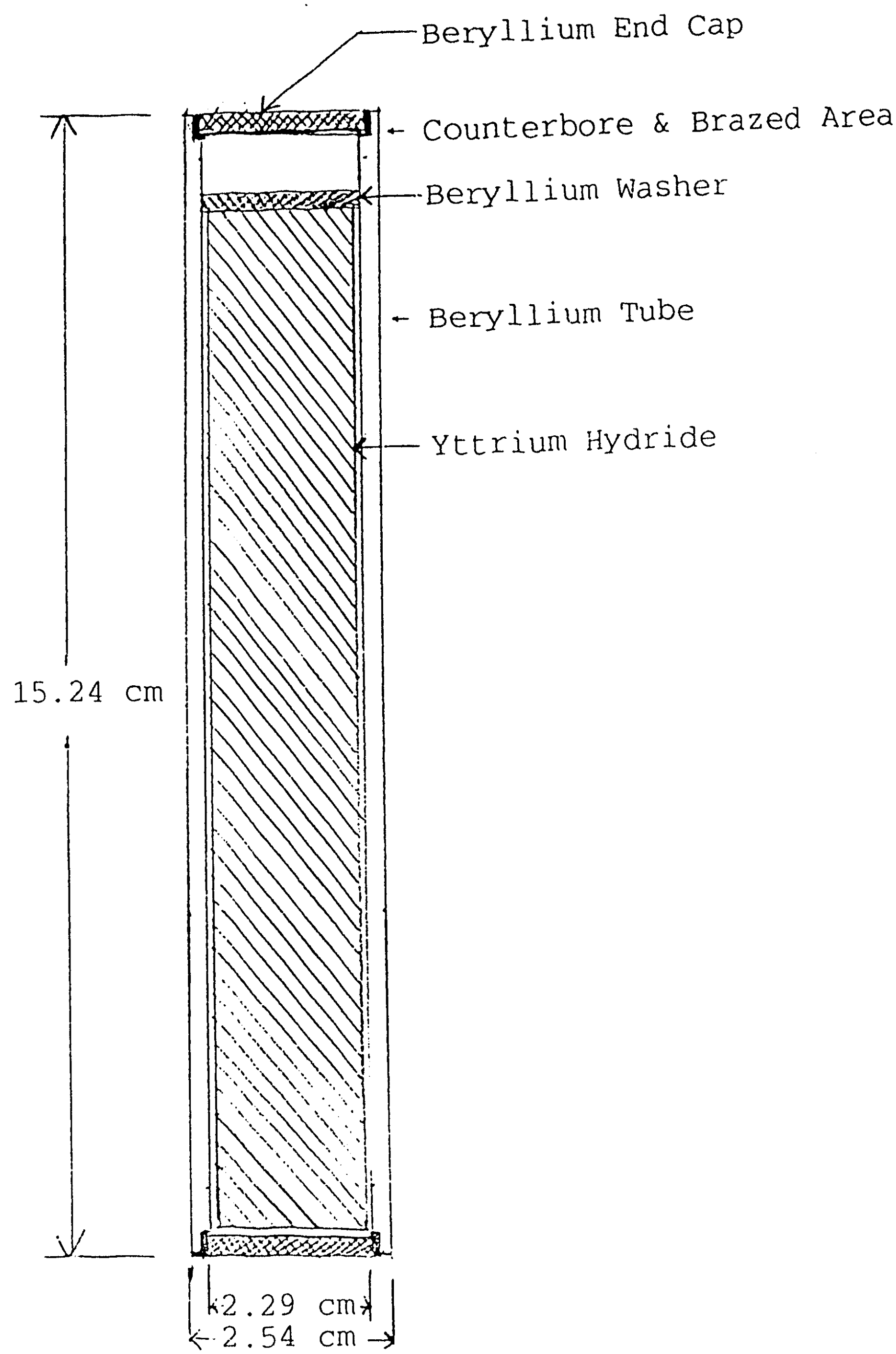



FIGURE 3. Yttrium Hydride - Beryllium Specimen With Endcaps Joined By Furnace Brazing

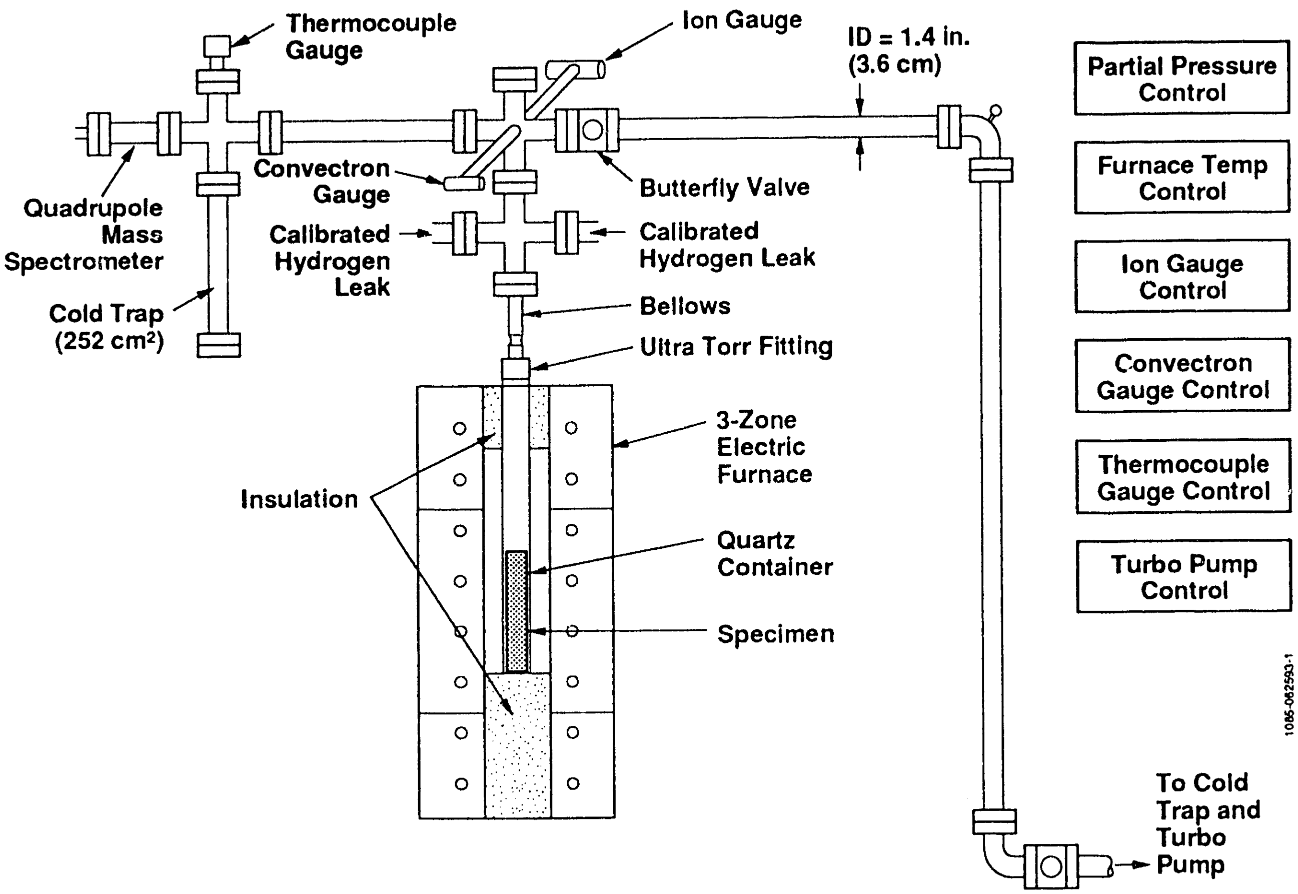



Figure 4. Experimental Setup for the Hydrogen Permeation Rate Measurement

Mod Test Schedule

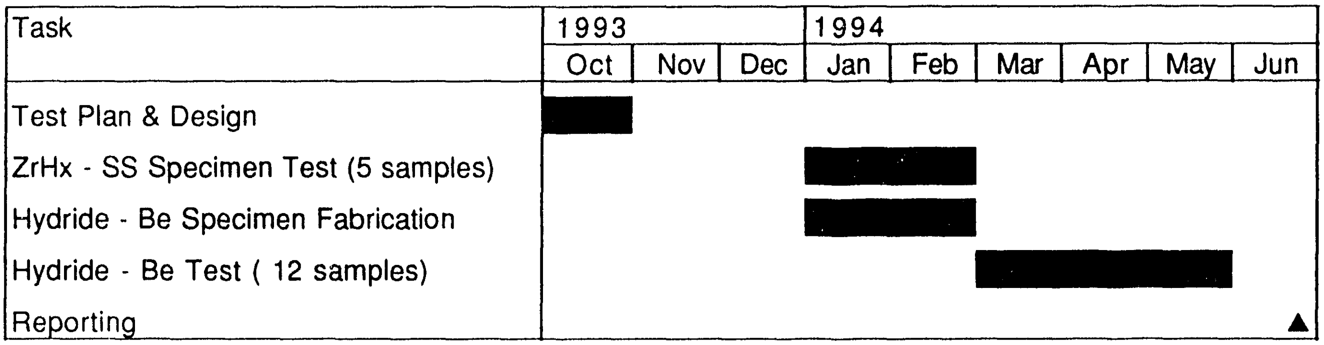



Figure 5. Moderator Containment Test Schedule

Unit : $1000's

| Task | GFY94 | | | Total |
|---|---|---|---|---|
| | 1Q | 2Q | 3Q | |
| Test Plan & Design | 10 | | | 10 |
| ZrHx - SS Specimen Test (5 samples) | | 33 | | 33 |
| Hydride - Be Specimen Fabrication | | 54 | | 54 |
| Hydride - Be Test ( 12 samples) | | 26 | 44 | 70 |
| Reporting | | | 6 | 6 |
| Total | 10 | 113 | 50 | 173 |

Notes :

1. Amounts are loaded cost
2. Cost for Hydride - Be test is $140K
3. Cost for ZrHx - SS test is $33K

Figure 6. Cost Breakdown