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ABSTRACT

Seven irradiated tritium-bearing lithium-bonded capsules containing vanadium alloy specimens were disassembled in a hot-cell facility at Argonne National Laboratory. These capsules were irradiated in the Fast Flux Test Facility as part of the structural materials research for the Fusion Power Program. The principal concern in disassembling these capsules to retrieve the test specimens was the hazard associated with the residual bonded tritium, which ranged from ≈17 to 99 Ci per capsule. This tritium had to be contained to the maximum extent possible. An enclosed system was built in which we used liquid ammonia to dissolve the lithium bond, alcohol to rinse and clean the retrieved specimens, and chemical getters (containing a reactive metal alloy) to trap the released tritium gas. The main processing units of the system are inside a hot cell and the remainder are in a vented cabinet. Essentially all of the tritium in the capsules was either trapped in the getters or confined in the solid reaction residues in beakers of spent ammonia and alcohol. There was no personnel contamination or accidental tritium release during the entire procedure.

INTRODUCTION

Owing to the desirable properties of low activation and high surface-heat-load capacity, vanadium alloys[1-3] are being considered as candidate materials for first wall/blanket applications in fusion reactors. One important issue to be addressed for this application is the effect of helium generated from vanadium (n,α) reactions in the 14-MeV neutron fusion environment. The presence of helium in the alloy structure can potentially affect physical and mechanical properties, such as density, strength, and ductility, of the material. Irradiation testing of vanadium alloys in fast fission reactors, such as the Fast Flux Test Facility (FFTF), typically produces <1% of the expected helium because of the lower energy of the fission neutrons. Therefore, to study the effects of helium generation, an innovative experimental technique was necessary. In the dynamic helium charging experiment (DHCE)[4] in the FFTF, this was achieved by doping the lithium bond of the specimens with tritium during fabrication. During irradiation (at 400-600°C), some of the tritium would diffuse into the vanadium alloy specimens and decay in situ into ^{3}He, thereby yielding the concurrent effects of neutron damage and helium generation.

The DHCE consisted of seven capsules, all 9.53 mm in diameter, 121 mm long, and made of the molybdenum-base alloy TZM. The lithium bond (≈1 g in each capsule) and the specimens (miniature tensile samples and transmission electron microscope disks) occupied approximately half of the internal volume; a gas plenum, to accommodate the helium gas from tritium decay, occupied the other half. The capsules were doped with various amounts of tritium. To offset tritium loss through the capsule wall, the lithium bond was enriched with ^{6}Li for compensatory tritium generation during irradiation. The tritium remaining in the capsules at the time of disassembly was estimated to be between 17 and 99 Ci per capsule.

The central requirements in disassembling the DHCE capsules were to preclude contamination of personnel and minimize tritium release through the exhaust stack. To satisfy these requirements, an enclosed system was built and used. The induced radioactivity in the TZM material dictated that at least the initial operations be conducted remotely in a hot cell.

This paper describes the processes, procedures, and equipment used to retrieve clean specimens while containing the tritium.

DESCRIPTION OF THE DISASSEMBLY SYSTEM AND PROCEDURE

Based on our previous positive experience of processing low-tritium capsules of similar design, we adopted the same basic approach for the present task, namely, using

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liquid ammonia to dissolve the lithium bond and alcohol to rinse and clean the retrieved specimens. It was concluded that an enclosed system was necessary to trap the released tritium gas. The enclosed system built for this task consisted of a capsule puncture jig to vent the high-pressure plenum gas that contained tritium, a chamber for ammonia dissolution and alcohol rinsing, a set of cold traps at liquid-nitrogen temperature to remove (by condensation) ammonia and alcohol vapors from the process gas stream, and chemical getters (containing a reactive metal alloy) to trap the tritium gas that was released during the dissolution and rinsing steps. The system was purged with high-purity (99.999%) nitrogen that carried tritium from the puncture jig and gaseous reaction products from the dissolution chamber through the traps and getters. The cleaned purge gas was then exhausted through the hot-cell stack.

The capsule puncture jig and the dissolution chamber were inside the hot cell and were operated remotely; the rest of the system, except the supply gas and liquid cylinders, was in a nearby vented cabinet and accessible through transparent sliding doors. A schematic diagram of the system is shown in Fig. 1 and the photographs of the components in the cell and in the vented cabinet are shown in Figs. 2 and 3, respectively. Except during capsule loading and unloading, both the in-cell and out-of-cell systems were isolated from the atmosphere and maintained under a nitrogen purge to minimize moisture in the system. Although the principal purpose of the sealed system was to contain the tritium, it also protected the operators from the potential chemical hazards of the ammonia gas.

A low-level beta-gamma hot cell in the Irradiated Materials Laboratory at ANL was chosen for this operation. The cell, 1.8 m wide x 1.5 m deep, has a movable front shield door with a zinc bromide-filled window for viewing and a pair of master-slave manipulators. All in-cell components for this task were installed through the shield door. Transfer ports and feed-through in the cell walls provided access for gas and liquid lines, electric cables, insertion of supplies, and removal of the clean test specimens. All large exposed surfaces of the cell interior were covered with polyethylene sheeting for ease of postoperation cleanup. The cell contains an air atmosphere and an exhaust fan maintains the cell at ≈0.9 mm Hg negative pressure when the door is closed.

The first step in the disassembly operation was to puncture the capsule to vent the high-pressure plenum gas. This was done in an enclosed jig that consisted of a cylindrical tube, a pressure gauge, and a modified high-pressure valve whose bore had been enlarged to accept the capsule and the valve stem fitted with a sharpened tip to act as the piercer. Prior to puncturing, the jig was thoroughly purged to remove the air introduced during loading of the capsule. Because the plenum gas contained tritium, the gas was gettered before being released.

After the puncturing, the capsule was removed from the jig and the ends were cut open to expose the contents for the subsequent dissolution and rinsing operations. This cutting operation was performed remotely in the air atmosphere of the cell with a conventional tubing cutter. Because essentially all of the tritium in the solidified lithium bond was in the stable chemical form of LiT and very little tritium gas release was expected, an enclosed system for this operation was deemed unnecessary. The tritium monitor in the cell exhaust stack confirmed that this was indeed the case. The opened capsule was then placed inside a wire-mesh basket and loaded into the dissolution chamber.

The dissolution chamber was by far the most critical component for successful operation. After extensive preoperational trials and design improvements, the finished unit incorporated a counterbalanced top cover capable of maintaining hermeticity up to $\approx 3 \times 10^4$ Pa (5 psi) positive pressure, a transparent Lucite (for viewing purposes) chamber body that could be slid back and forth to facilitate removal/replacement of beakers of ammonia and alcohol in which the dissolution and rinsing were carried out, a sealed mechanism to lower or raise the specimens (in the wire-mesh basket) into or out of the ammonia or alcohol liquid, a feed line that permitted the loading of liquid ammonia (b.p. -33°C) directly from a supply cylinder without breaking the hermeticity, a heater in the base to evaporate the excess liquid ammonia at the end of the dissolution process, and all of the necessary purge/carrier gas lines.

After purging to remove the air introduced during loading, the basket was gradually lowered into a beaker of liquid ammonia in the sealed chamber to begin the dissolution process. The tritium gas generated during the dissolution process (along with the evaporated ammonia) was driven from the dissolution chamber by the nitrogen carrier gas to the cold traps and then to the tritium getters. The cold traps, the purpose of which was to eliminate the ammonia vapor from the gas stream, were commercially-available, thick-wall Type 304 stainless steel cylinders, 680 mm long, 102 mm in diameter, with reduced-diameter, threaded ends. During operation, the middle ≈400-mm section of the cylinder was chilled with liquid nitrogen; the ends were intentionally not chilled to avoid plugging of the gas passage by solidified ammonia (m.p. -78°C). Three cylinders were used as cold traps: two in parallel as the first stage and a third as the second stage. Bypass lines for each stage were provided to circumvent plugging in an individual cold trap, but were never needed for that purpose.

The relatively large chilled surface area inside the cylinders, the low vapor pressures of ammonia and alcohol at the liquid-nitrogen temperature, and the long dwell time of the gas in the cylinders contributed to excellent trapping efficiency, even without the aid of adsorbent materials, such as molecular sieves, in the cylinders. (Sieves were undesirable because of the possibility of off-gassing which could damage the tritium getters and because of difficulties with postoperation disposal.) The high pressure rating of the cylinders (12.4 MPa) permitted temporary storage of trapped ammonia (0.91 MPa vapor pressure at room temperature) until later, when it was time for venting.

Tritium trapping was accomplished with two, and occasionally three, commercially available getters* arranged in series. The getters contained sintered Zr-Fe powders and provided a near 100% trapping efficiency as long as the capacity was not saturated. Because the getters react with all impurity gases (e.g., oxygen, ammonia, and alcohol), and can become significantly overheated if the reaction rate is excessive, particular care was taken to protect the capacity and, more importantly, the integrity of the getters. Thorough purging was performed each time before trapping to remove the air introduced into the system during loading of specimens or changing of beakers.

Following the ammonia dissolution, the specimens (still in the wire-mesh basket) were cleansed with alcohol in a new beaker in the dissolution chamber to remove the surface residues. The procedure was essentially the same as described above for ammonia dissolution. Although the quantity of tritium release during this phase of the operation was greatly reduced, both the cold traps and getters were used to minimize the tritium effluent.

A second alcohol rinse in another clean beaker was then performed to prepare the specimen for discharge from the cell. Because the risk of tritium release at this point was small, this step was conducted in the open atmosphere of the cell without the tritium getters.

OPERATING EXPERIENCE

The puncturing jig and the dissolution chamber were both designed and built in-house. In designing these units, considerable effort was spent to achieve rugged construction and easy remote operation. These efforts and the extensive preoperational trials resulted in excellent performance characteristics in both units. No maintenance was required on either unit after activation.

*SAES Getter/USA Inc., Colorado Springs, CO.

The irradiated TZM capsule material was brittle and tended to fracture longitudinally when being opened with the tubing cutter. The tendency to fracture was actually advantageous because it exposed more lithium surfaces to ammonia dissolution. Typically, the lithium bond would be completely dissolved in <2 h. The entire processing of a capsule, including the time for interim cell cleanup, required approximately five working days.

The temperature of the cold traps varied between -196°C (b.p. of liquid nitrogen) and ambient. An ordinary epoxy was initially used to secure the threaded gas line fittings onto the cold trap cylinders. This proved to be unsatisfactory because leakage would sometimes develop, apparently due to temperature cycling. This problem was eliminated by switching to an epoxy designed for cryogenic applications.

Flow meters, pressure gauges, thermometers, and tritium monitors were used at various locations in the system (see Fig. 1) to provide operational data. More importantly, these sensors gave advanced warning of potential troubles to permit remedial actions.

In an early dry run with no tritium in the system, the second (i.e., downstream) tritium getter overheated and resulted in the burning of the insulation on its power cables. The cause was a portable tritium monitor that had been positioned between the two getters to monitor the performance of the first getter. The monitor had a built-in recirculating pump to sample the process gas in the line. Apparently, the recirculating-pump system had a small air leak (which could not be detected during the system leak check) that introduced oxygen into the line and caused the excessive reaction with the getter alloy. This situation was rectified by relocating the tritium monitor downstream of all getters. After this modification, and with enhanced awareness of the need for thorough purging, no further overheating incidents were encountered in subsequent operations.

In addition to the aforementioned portable tritium monitor and the monitor already in the cell exhaust duct, two additional detectors (ionization chambers) were installed in the system to monitor the tritium concentrations in the process gas. Originally, the carrier gas designed for the system was high-purity helium. During trial runs, however, it was found that helium gas would cause the two ionization chambers to arc and malfunction. The same was true with argon gas. Because sending the already-contaminated tritium monitors back to the vendor for modification was out of the question, we decided to use high-purity nitrogen as the carrier gas to circumvent the arcing problem. Because nitrogen is not as inert as he-

lium, it is likely that the switch to nitrogen sacrificed some of the capacity of the tritium getters. Possibly because of this, the getter cartridges required replacement twice during the procedure to provide adequate capacity. Replacement of the tritium-bearing reactive-metal cartridges in a fume hood, however, was straightforward and caused no contamination.

Another problem was associated with the two tritium monitors. Apparently, when the tritium concentration in the process gas exceeded the full-scale range of the monitors (200 Ci/m^3, which was reached on two occasions with the highest tritium capsules), the field-effect transistors (FETs) in the monitors would fail. On-site repair to clean the chamber and to replace the failed FETs was thus necessary. Fortunately, these repairs, also performed in a fume hood, resulted in no personnel or hood contamination.

All flow meters were calibrated beforehand with nitrogen, ammonia, and various combinations of nitrogen/ammonia flows. The substantial boil-off of liquid ammonia during the initial phases of the dissolution operation caused an increased flow and a noticeable back pressure in the dissolution chamber. The readings of the back pressure and the combined flow rates at various points in the system provided signature-like indications of the performance of the system, including the leak-tightness of the entire system and the efficiency of the cold traps. The use of sensors at key locations in the system proved to be extremely beneficial.

The reaction residues in the spent beakers of ammonia contained a substantial fraction of the total tritium inventory from the capsules. After a capsule was processed and the residual ammonia evaporated, each spent beaker of ammonia was placed in a gallon-size sheet metal can and temporarily stored in the hot cell. These beakers are awaiting disposal of as stabilized solid waste. The spent beakers of alcohol are stored in the same manner, though their tritium content is significantly lower.

Because tritiation of ammonia was negligible, it was possible to vent the condensed ammonia in the cold traps directly through the cell exhaust after each capsule was processed. Venting was accomplished under approved meteorological conditions and in a fully monitored manner. The quantities of ammonia vented from each trap (i.e., nearly one-half of the total in each of the two first-stage units and essentially nothing in the second-stage unit) provided a good indication of the efficient performance of the cold-trap system.

CONCLUSIONS

All seven tritium-bearing capsules in the DHCE series were processed and the specimens were successfully cleaned and retrieved. Based on data from tritium monitors, essentially all of the tritium in the capsules was either trapped in the getters or confined in the solid reaction residues in the beakers of spent ammonia. No personnel contamination or accidental tritium release occurred during the entire procedure.

The capsule puncture jig and the dissolution chamber specifically design for this task performed as expected during the entire procedure. The only notable difficulties were those encountered with tritium monitors. Arcing in the monitors necessitated the use of nitrogen, instead of helium or argon, as the process gas. This substitution possibly decreased the capacities of the tritium getters. Failure of the FETs in the monitors due to signal saturation required field repairs. The problems associated with the tritium monitors will have to be rectified before the system is used again for future DHCE-type experiments.

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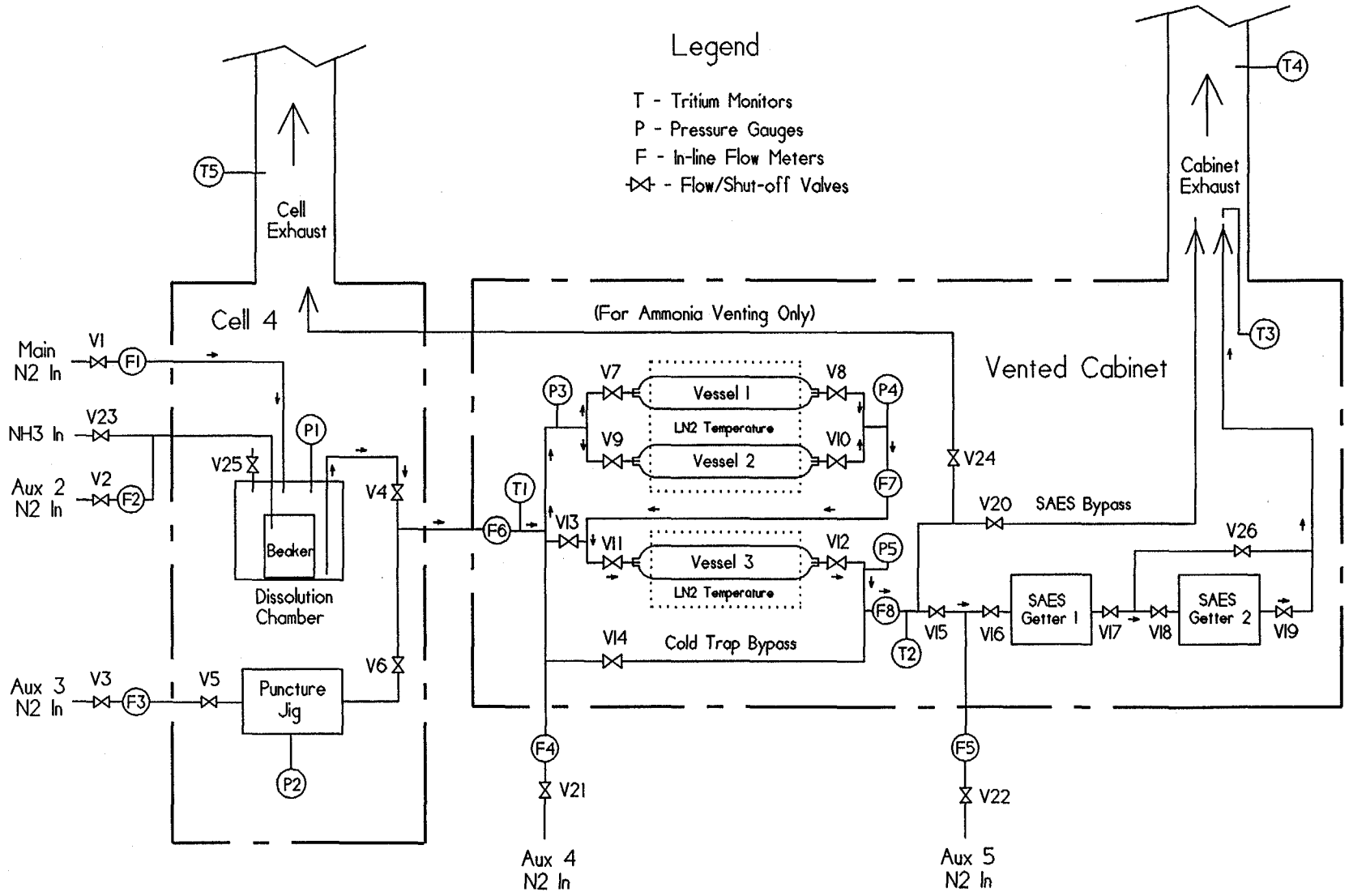

Figure 1. Schematic diagram of DHCE capsule disassembly system.

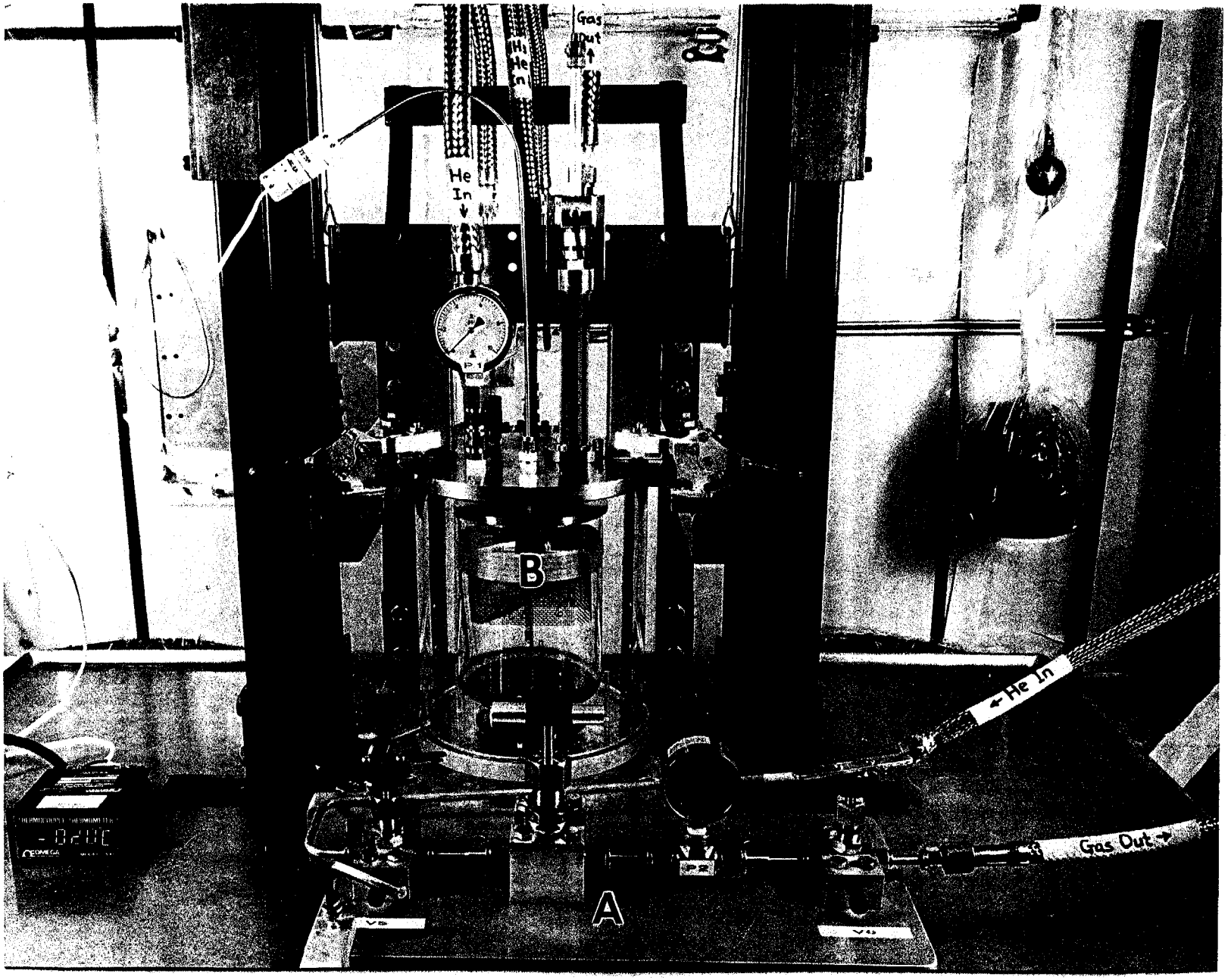

Figure 2. DHCE capsule disassembly system in cell, consisting of capsule puncture jig (A) and dissolution chamber assembly (B) (ET284650).

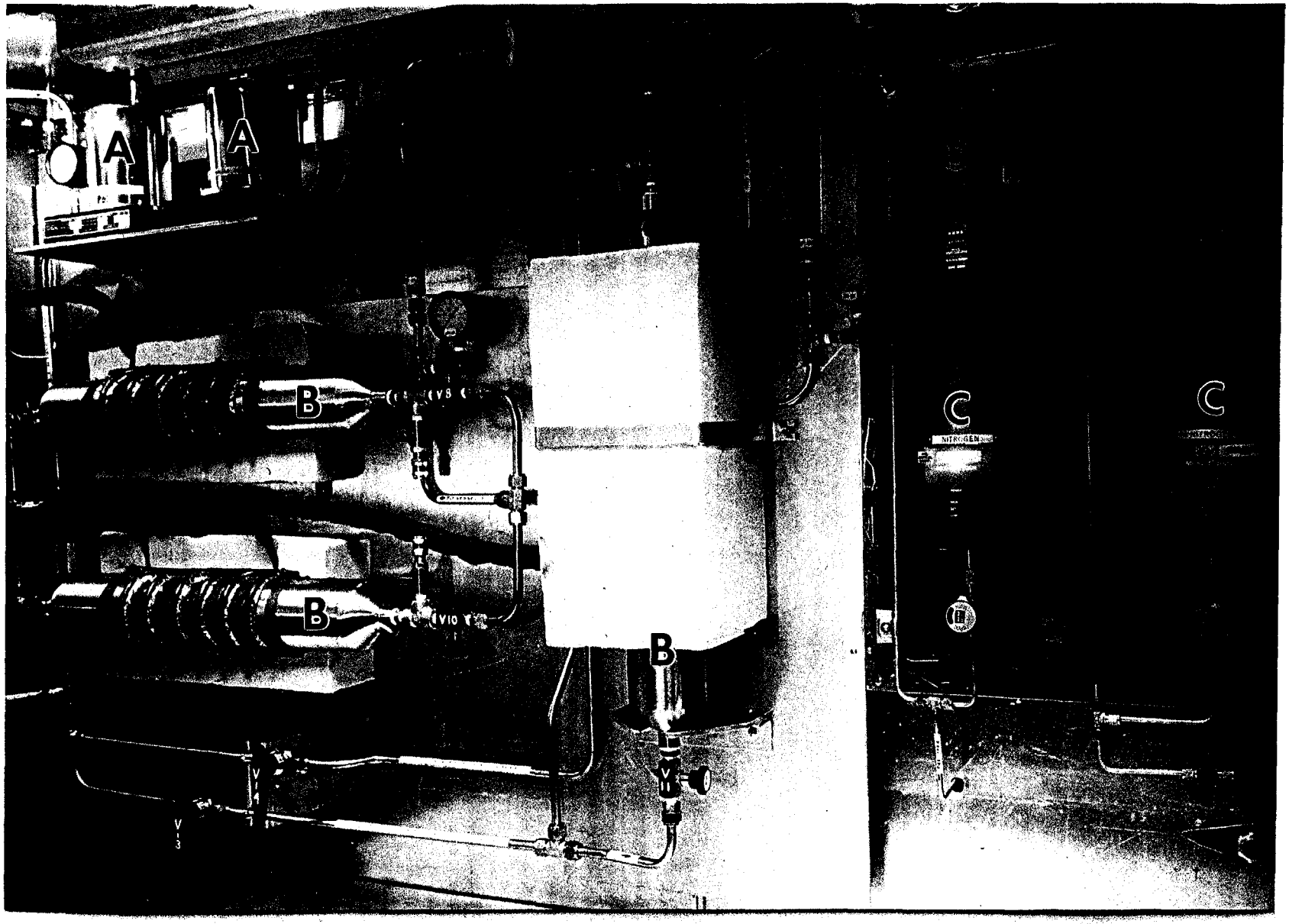

Figure 3. DHCE capsule disassembly system in the vented cabinet consisting of two tritium monitors (A), three cold traps, or ammonia condensers (B), and two tritium getters (C) (ET284651).