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THE INTEGRAL FAST REACTOR FUELS REPROCESSING LABORATORY AT ARGONNE NATIONAL LABORATORY - ILLINOIS

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INTRODUCTION

The processing of Integral Fast Reactor (IFR) metal fuel utilizes pyrochemical fuel reprocessing steps. These steps include separation of the fission products from uranium and plutonium by electrorefining in a fused salt, subsequent concentration of uranium and plutonium for reuse, removal, concentration, and packaging of the waste material. Approximately two years ago a facility became operational at Argonne National Laboratory-Illinois to establish the chemical feasibility of proposed reprocessing and consolidation processes. Sensitivity of the pyroprocessing melts to air oxidation necessitated operation in atmosphere-controlled enclosures.

INERT ATMOSPHERE GLOVE BOXES

The IFR Fuels Reprocessing Laboratory includes two high purity (<1 ppm O_2, <1 ppm H_2O, <10 ppm N_2) argon atmosphere glove boxes which are maintained at negative pressure for plutonium containment. Figure 1 shows a glove box and its associated purification equipment. The glove box design has been used successfully for maintaining high purity atmospheres or for containment of plutonium activity. The glove boxes are constructed of steel and each module contains laminated safety glass windows which have provisions for gloves. Thirty mil thick Hypalon gloves purchased from the Charleston Rubber Company, Charleston, SC, are used throughout. Each glove box has an 8-in. bag port and a 22-in. bag port which are shown in Figure 2; bagging techniques are used to transfer equipment into the glove

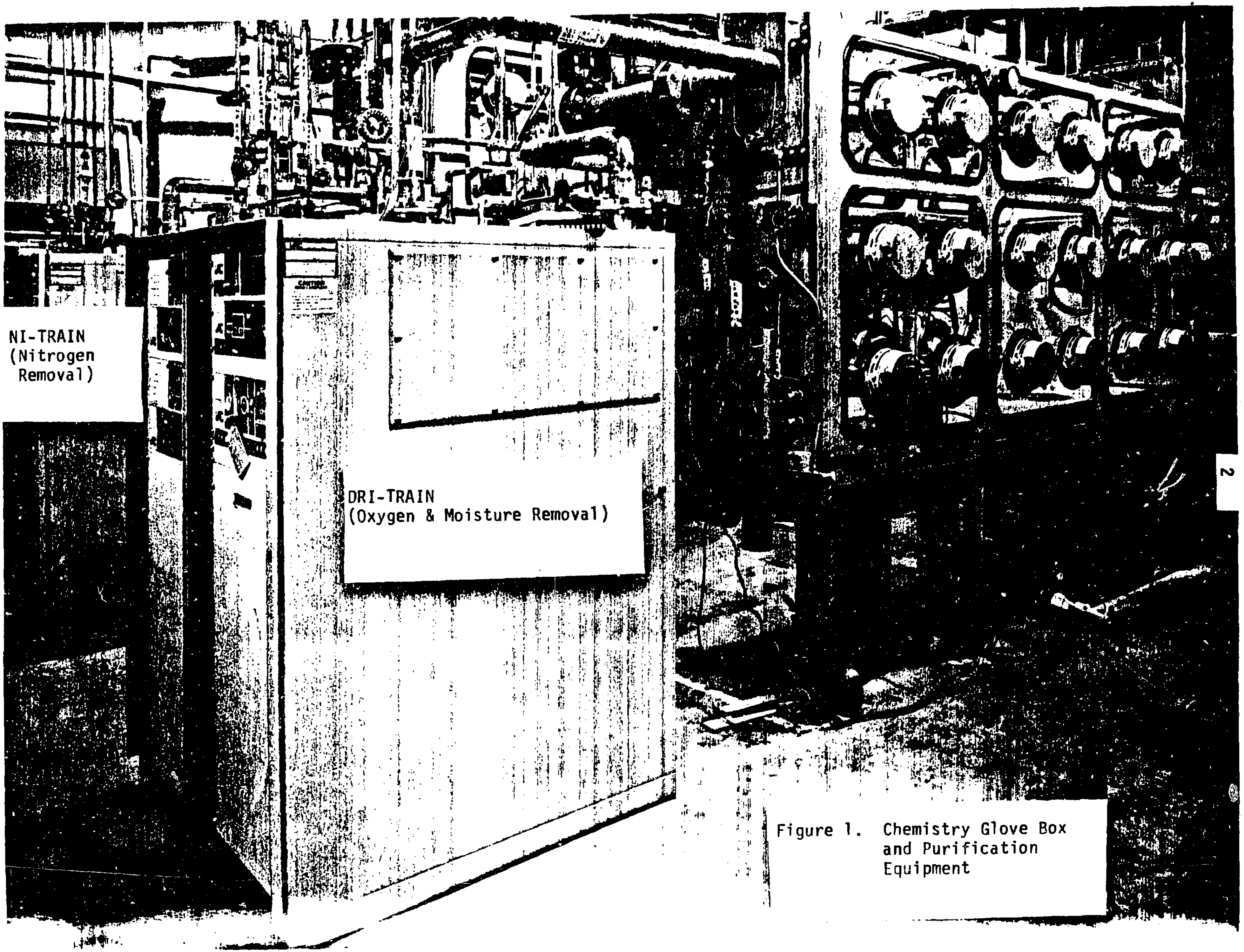

Figure 1. Chemistry Glove Box and Purification Equipment

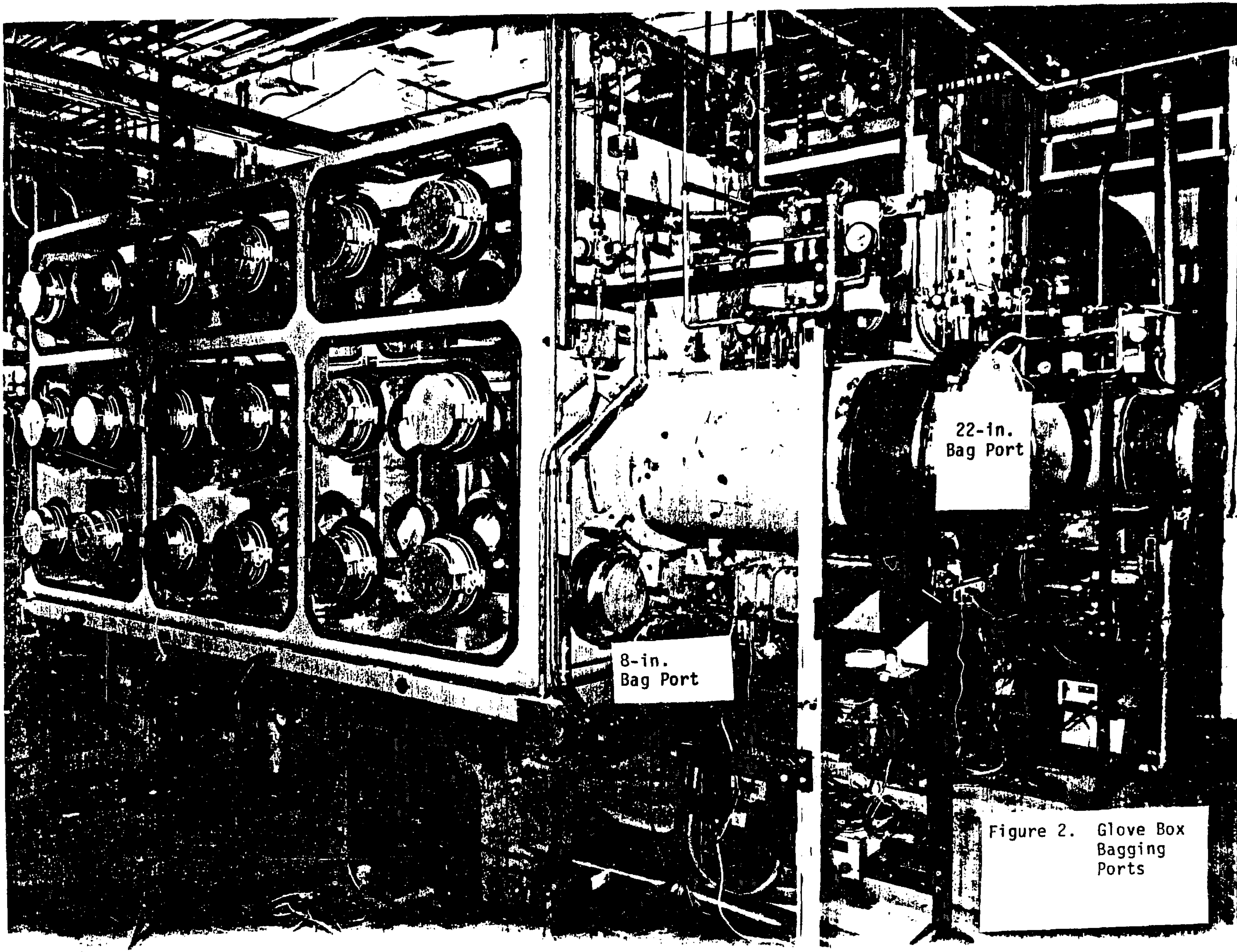

Figure 2. Glove Box Bagging Ports

box. Separate purification systems service each glove box and continually remove oxygen, moisture, and nitrogen from the argon atmospheres as well as maintain glove box pressure in the range of -0.4 in. to -1.2 in. H_2O. The moisture and oxygen levels in each glove box are continuously monitored, while the nitrogen level is measured on a need basis, usually once a day.

Each glove box is provided with an emergency pressure control system which protects the glove box against catastrophic failure in the case of a pressure rise to ≥-0.15 in. H_2O or a pressure reduction to ≤-2.0 in. H_2O. An abnormal pressure condition must persist for at least three (3) seconds before the emergency system is activated. This three-second requirement is necessary to prevent triggering the system through normal glove box operations.

Two glove boxes are currently operational in the laboratory. One glove box is designated for laboratory-scale (≤10 g Pu) experiments to investigate process chemistry. The second glove box accommodates engineering-scale (>10 g Pu) experiments.

CHEMISTRY GLOVE BOX

The glove box in which process chemistry experiments are performed is three modules long, one and one-half modules high, and one module deep. Dimensions of the enclosure are 3.03 x 1.60 x 1.06 m (10 x 5.25 x 3.5 ft) which provides a volume of 5.1 m^3 (183.8 ft^3).

Four resistance-heated furnace wells are attached to the glove box floor and become part of the glove box enclosure. These furnace wells provide experimental temperature capability to 700-800°C. Three identical wells have dimensions of 12.06 cm (4.75 in.) diameter and 51.5 cm (20.25 in.) deep, while the fourth well has a 25.4 cm (10 in.) diameter and is 75.57 cm (29.75 in.) deep. Instrumentation and power connections as well as piping connections for inert gas and vacuum for use in the glove box pass through penetrations in the glove box walls. Solution stability, rate of dissolution, and small-scale electrorefining experiments have been performed in this glove box.

ENGINEERING GLOVE BOX

The engineering glove box is three modules long, two modules high, and one module deep. Dimensions are 3.03 m x 2.06 m x 1.06 m (10 x 6.75 x 3.5 ft) providing a volume of 6.62 m^3 (236.25 ft^3). This glove box accommodates engineering-scale (>10 g Pu) experiments. An engineering-scale electrorefining cell and an induction furnace for product consolidation are located in this larger glove box as shown in Figure 3. As in the Chemistry glove box instrumentation and power connections as well as piping connections for inert gas and vacuum for use in the glove box pass through penetrations in the glove box walls.

ENGINEERING GLOVE BOX EQUIPMENT

A. Electrorefining Cell

The engineering-scale electrorefining cell, schematically shown in Figure 4, is located in the engineering glove box. A 25.4 cm (10 in.) diameter x 75.57 cm (29.75 in.) deep furnace well is mounted to the floor of the glove box in the center module. This well serves as an attachment point for a flange used as a support ring from which the crucible and liner are hung. The crucible, liner, and support ring are assembled and positioned in the furnace well as one unit. Figure 5 shows these components as

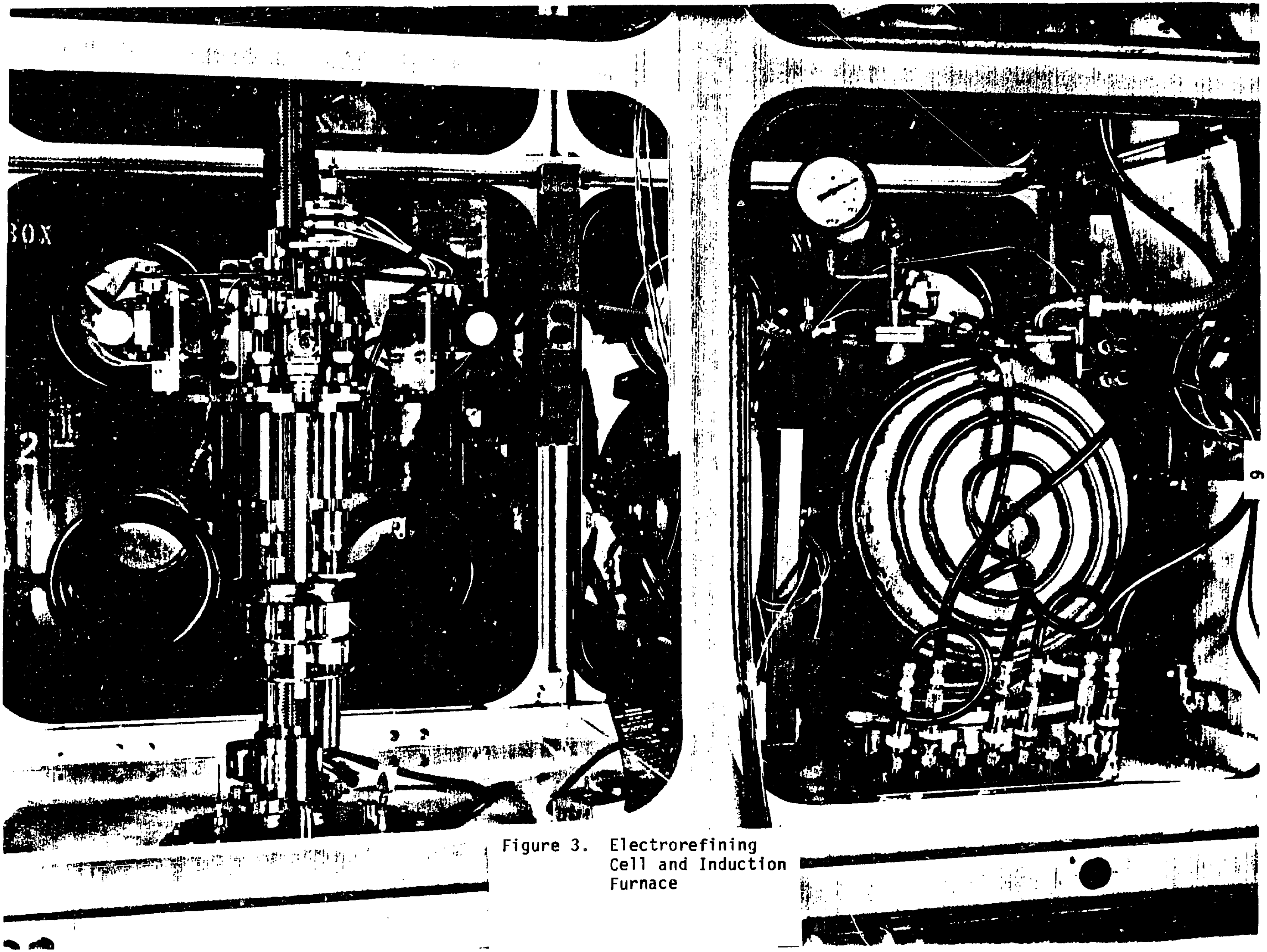

Figure 3. Electrorefining Cell and Induction Furnace

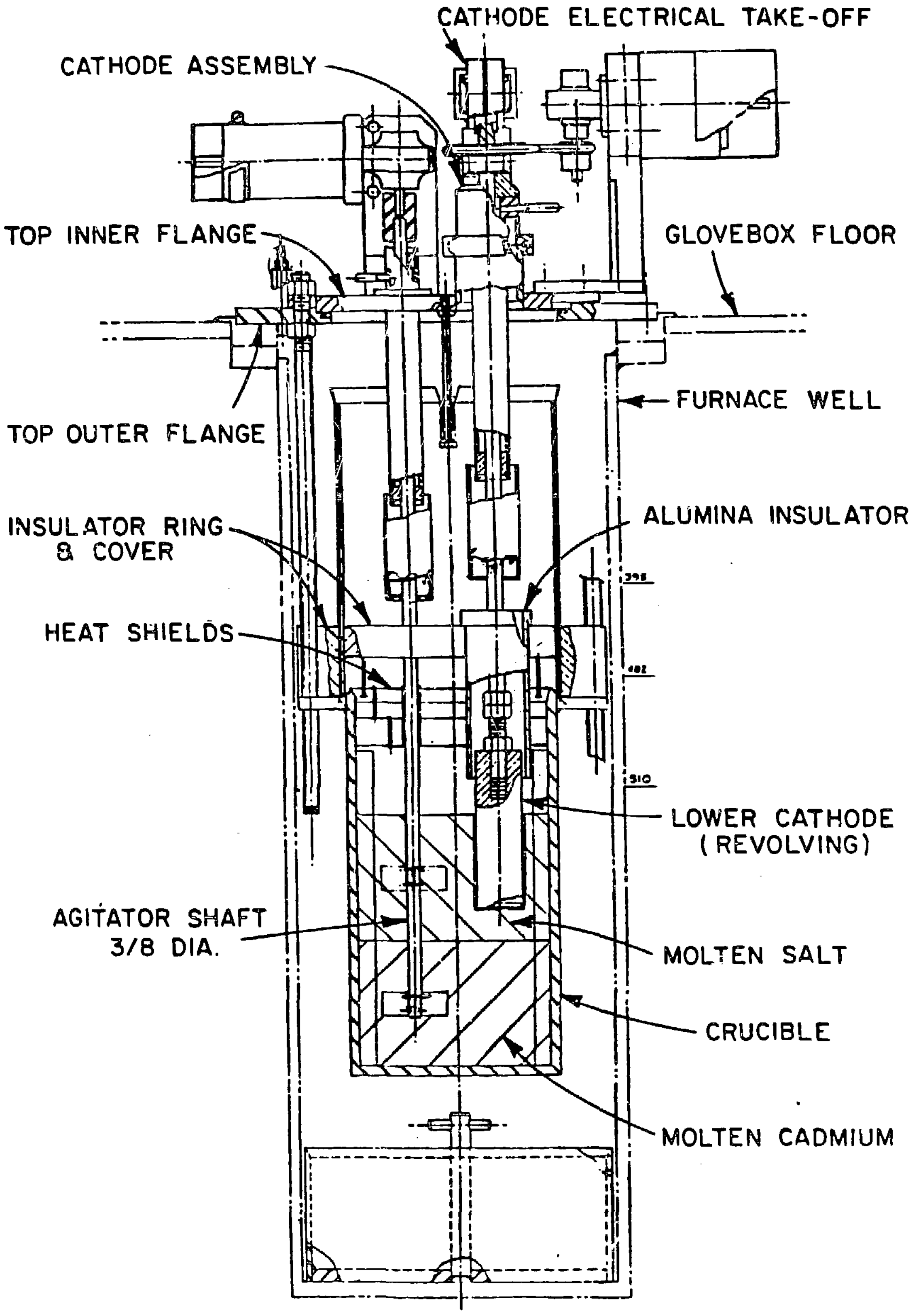

Figure 4. Engineering-Scale Electrorefining Apparatus

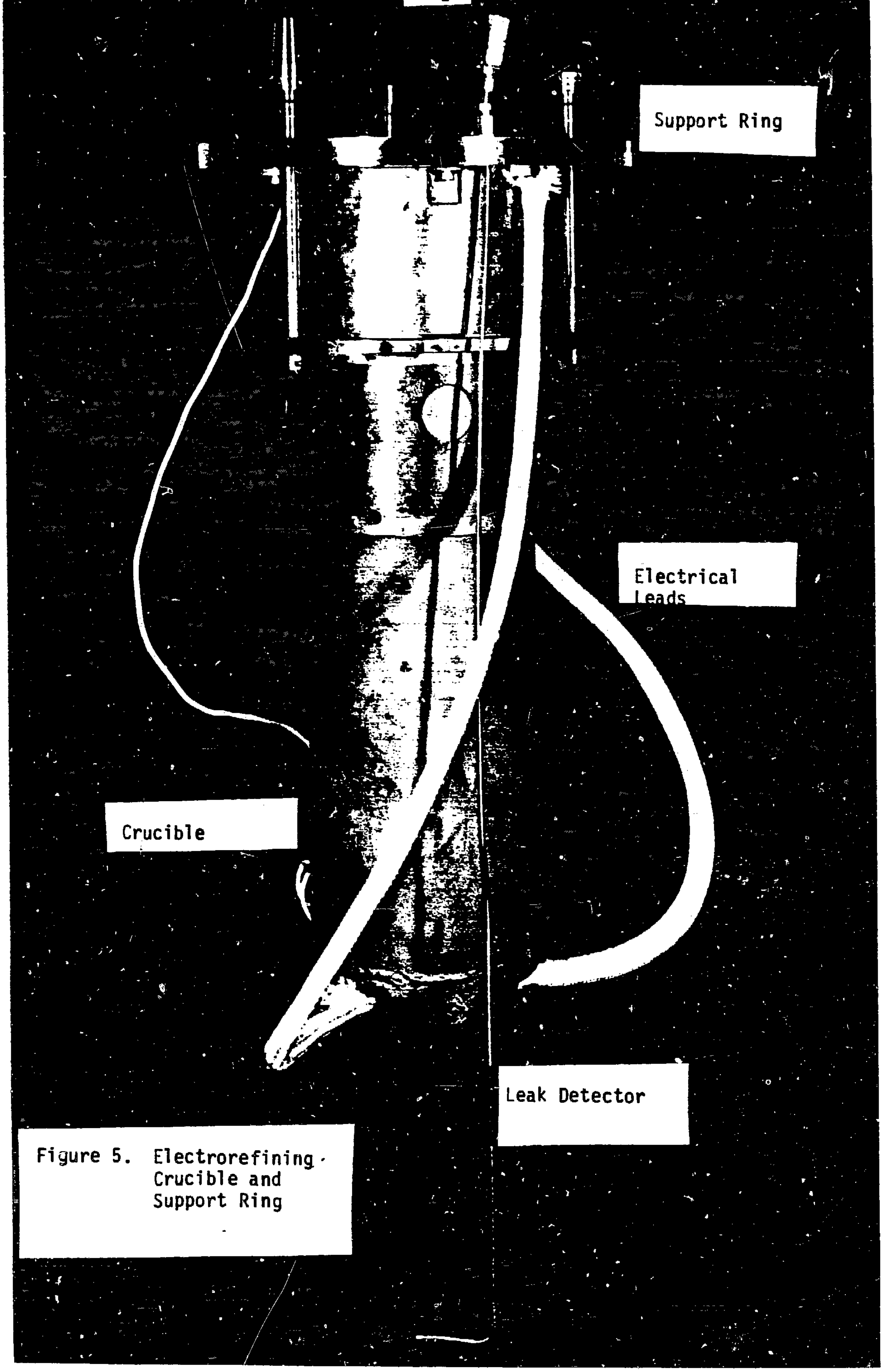

Figure 5. Electrorefining Crucible and Support Ring

well as electrical leads and a leak detector to signal leakage in the well. The crucible is constructed from mild steel and has a 16.25 cm (6 in.) diameter and is 33.8 cm (13.3 in.) deep. Three threaded rods passing through the support ring and pinned to the crucible allow the height of the crucible to be adjusted over a 12.7 cm (5 in.) range.

The upper flange assembly shown in Figure 6 has penetrations for the cathode housing assembly, agitator housing assembly, reference electrode, thermowell, and a ball valve. The complete Electrorefining Cell is shown in Figure 7. A cylindrical mild steel cathode is threaded onto the lower portion of the cathode bearing housing. Bearings in the cathode housing allow the cathode to rotate freely. A Bodine gearmotor* model 32D5BEPM-5F-142 and variable speed controller allow the cathode to be rotated up to 125 rpm. The depth of the cathode is adjustable over a 2.5 cm (1 in.) range by adjustment of a threaded rod on the cathode housing. In one successful cathode design a mild steel cylinder 3.175 cm (1.25 in.) diameter and 30.0 cm (11.8 in.) long has provision for a voltage probe to be positioned within 6.67 cm (2.625 in.) of the tip to monitor cell voltages. Shaped copper blocks mounted in a nylon holder are held against the hardened upper cathode shaft under tension to provide current pickups. A friction connection to the voltage probe is also made at this point. Fine adjustment of the cathode and crucible heights allows precise location of the cathode in the electrolyte.

*All motors used in the glove box have brushes fabricated from Stackpole Grade 605 carbon-graphite material containing MoS_2 for use in dry atmospheres.

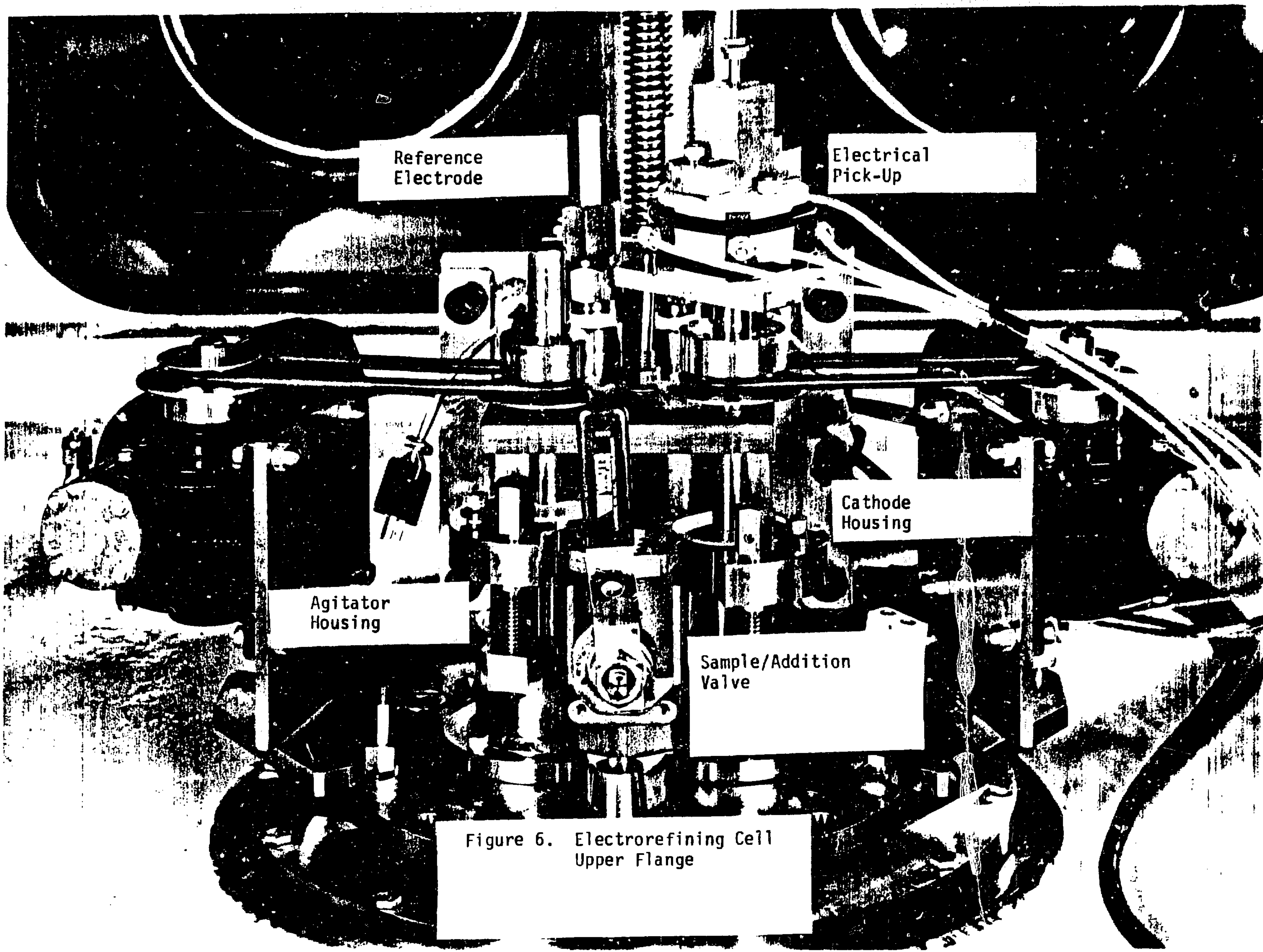

Figure 6. Electrorefining Cell Upper Flange

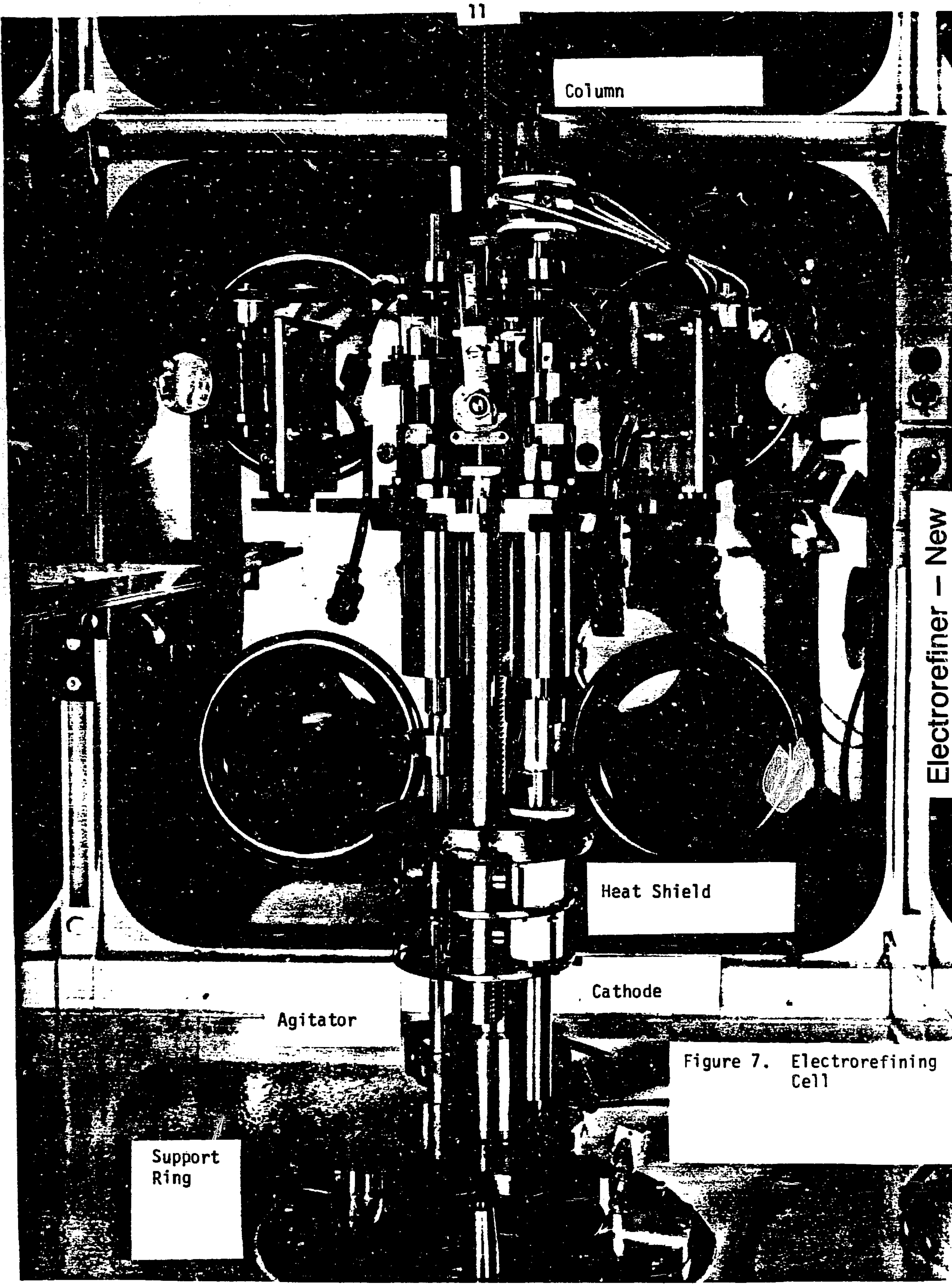

Figure 7. Electrorefining Cell

The mild steel agitator is a simple two-bladed stirrer with flat bladed paddles located in the electrolyte, the cadmium anode, and at the salt/metal interface. Location of the agitator shaft is also adjustable over a 2.5 cm (1 in.) range. It is necessary to position the agitator so that the electrolyte, metal phase, and interface are stirred simultaneously to aid mass transport. The agitator is rotated at speeds up to 300 rpm using a belt and pulleys driven by a Bodine gearmotor model 32D5BEPM-5F-141 and variable speed controller.

Conoseal fittings are used to secure the Ag/AgCl reference electrode and thermowell. The thermowell is a 0.64 cm (0.25 in.) diameter x 0.16 cm (0.063 in.) wall x 86.36 cm (34 in.) long mild steel closed end tube. A 2.5 cm (1 in.) diameter ball valve is also mounted on the upper flange of the electrorefining cell. This valve allows materials to be easily added to the crucible or the melt sampled.

The upper flange of the ER cell may be raised or lowered by a gearmotor which is mounted on a 7.62 cm (3 in.) diameter column 115.57 cm (45.5 in.) long bolted to the glove box floor. A 107.00 cm (42.125 in.) long threaded rod (1-5 ACME thread) is suspended from the column and the gearmotor mounted on top of the column drives the thread directly. A captive nut attached to the flange moves along the threaded rod to raise or lower the flange and components. A Bodine gearmotor model 32D5BEPM-5F-142 and variable speed controller allows smooth movement of the flange up or down. Limit switches are used to shut off the motor at the fully raised or full lowered positions. A keyway is fastened to the column to align the upper flange while raising or lowering. This keyway does not extend the full length of the column and allows rotation of the flange at its maximum raised position. The flange may be rotated into an open work area in the adjacent module for maintenance if necessary.

In normal operation a product such as shown in Figure 8 would be deposited on the cathode. This product is obtained from operation of the cell and requires that the entire assembly be raised in order to remove the cathode. To minimize heat buildup in the glove box, the furnace well is quickly covered with several baffles and a heat shield after the assembly is lifted out of the well. When the upper assembly is cool, the product is stripped off the cathode. The product contains dendritic metal and salt. The salt must be separated from the metal by heating to high temperature (~1250°C) in a BeO crucible using the induction furnace which is located in the adjacent module.

The charge to be heated is loaded into an isostatically pressed BeO crucible typically 6.35 cm O.D. x 5.08 cm I.D. x 12.38 cm high (2.50 in. x 2.0 in. x 4.875 in.). The crucible is located inside a graphite susceptor within the induction coil. An ATJ grade graphite shell packed with graphite felt is used as a fume trap to catch volatiles. A thermowell is located in the fume trap to monitor temperature in the vapor space. At the completion of the heating cycle, the melt is poured into a tapered ATJ graphite mold. The salt and heavy metal separate into two layers in the mold. A detent in the bottom of the mold allows a sample to be broken off the heavy metal ingot for analysis.

B. Induction Furnace

The 10 KHz induction furnace is located in the third module of the glove box. The stainless steel vacuum chamber has an inner diameter of 45.7 cm (18 in.) and is 53.3 cm (21 in.) deep. The inner diameter of the

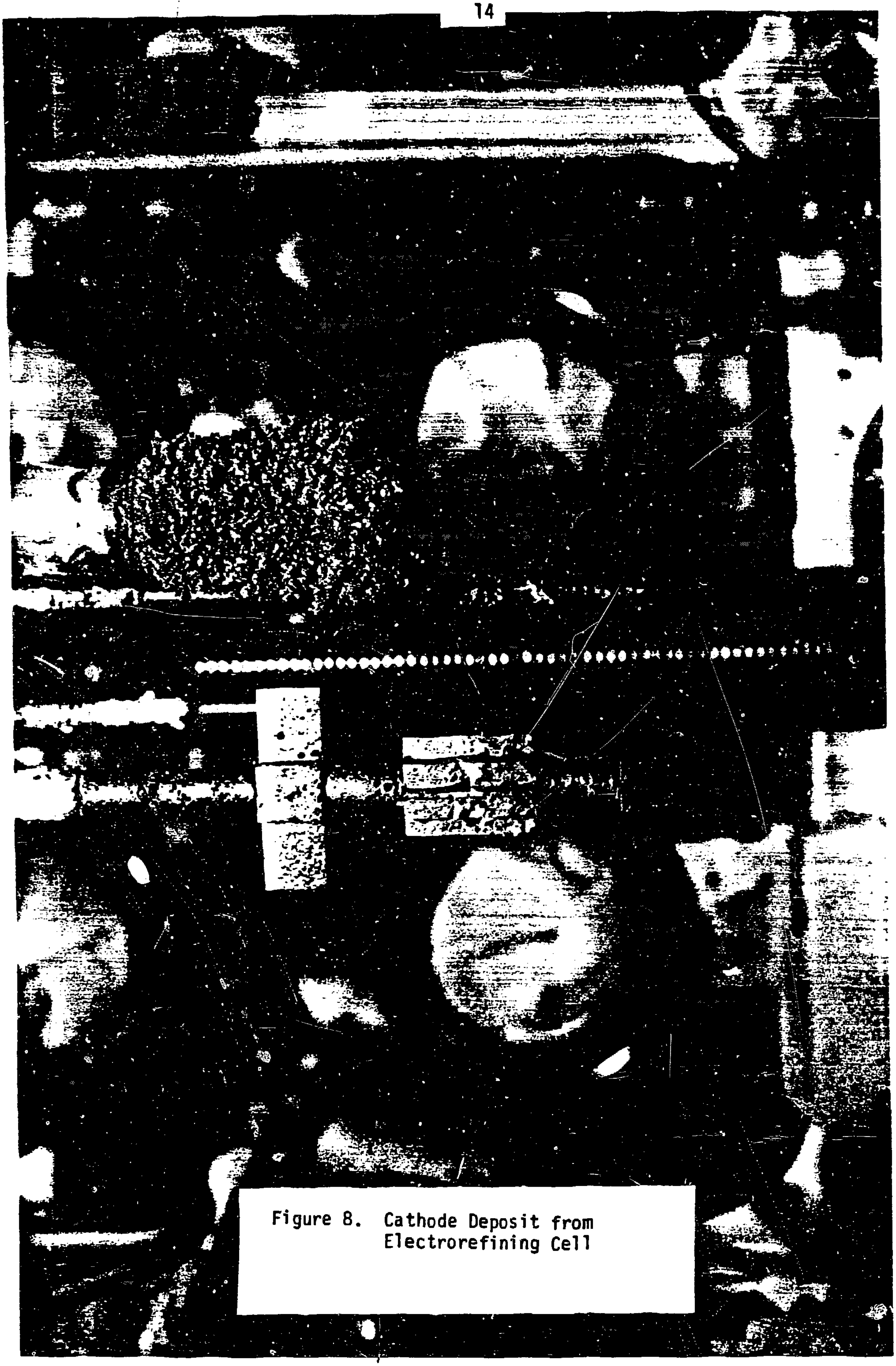

Figure 8. Cathode Deposit from Electrorefining Cell

base flange is 49.2 cm (19.4 in.). O-ring seals are used at the door and base. The chamber may be evacuated and pressurized to 725 mm with glove box atmosphere. Five cooling water circuits service the furnace; one for the door, three for the shell, and one for the base. The base elevates the shell 5.7 cm (2.25 in.) and allows a 34.3 cm (13.5 in.) diameter feed-through plate to bring the necessary supply and return lines for the cooling water circuits through the floor of the glove box. Continuous 1.27 cm (0.50 in.) diameter stainless steel tubes bring the cooling circuits into the box, and a transition is made to short lengths of pressure hose in the glove box by means of Swagelok Quick Connect fittings. During periods when the furnace is not to be used, the cooling lines are blown dry using argon. If more extensive maintenance were necessary, after the cooling lines are dry, the Quick Connect fittings may be undone and an effective moisture-tight seal to the glove box atmosphere maintained.

The coil assembly of the furnace is shown in Figure 9. Supporting the coil at both sides stabilized the assembly and allows easy tilting of the furnace even at temperatures above 1300°C. The coil is seven turns and is formed from rectangular copper bar whose dimensions are 0.95 x 1.27 cm (0.375 x 0.05 in.). The coil itself has an inner diameter of 12.7 cm (5.0 in.), is 13.55 cm (5.38 in.) high and is attached to the tilting mechanism. The coil can be easily removed and is replaceable if necessary.

Each cooling water circuit (six in all, one for the work station) has a flowmeter to provide visual indication of flow and a flow switch to provide system shut down if the volume of cooling water in that circuit falls below a predetermined level. When low water volume in a circuit occurs, the system alarms and the main water supply valve closes and

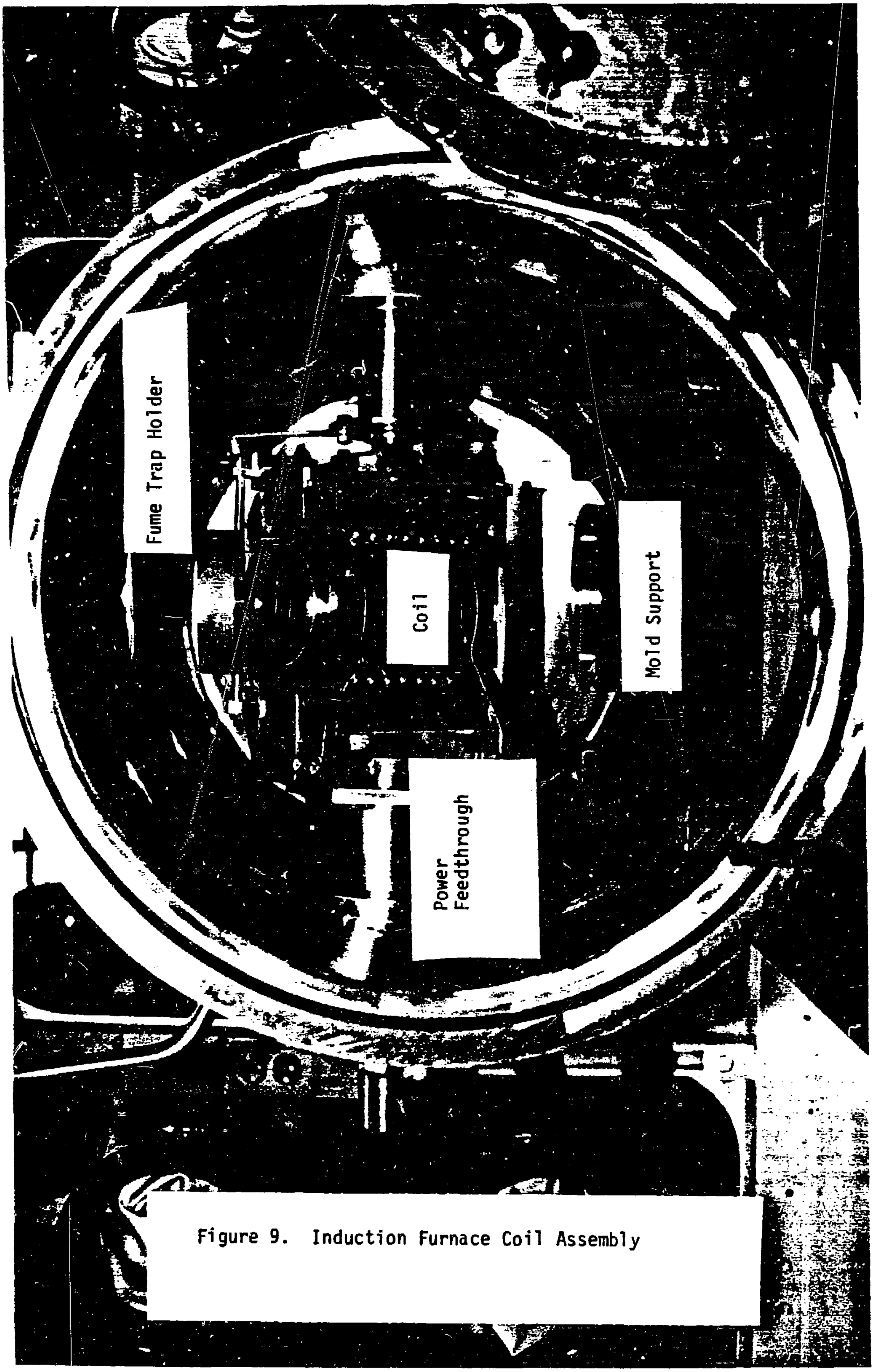

Figure 9. Induction Furnace Coil Assembly

remains closed until the situation is corrected and the system reset. A cooling supply shutdown can also be triggered by a high ppm moisture level in the glove box. This moisture level can be arbitrarily chosen by adjusting the moisture monitor. When this level is reached, the moisture monitor will indicate an alarm condition and cause shutdown until the condition is corrected and the system reset.

ATMOSPHERE PURIFICATION SYSTEMS

The argon atmosphere of each glove box is maintained at low levels of oxygen, moisture, and nitrogen by passing the box gases through commercial purification systems. Each glove box is serviced by a Model MO-120-2 DRI-TRAIN and a Model NI-40 NI-TRAIN manufactured by Vacuum/Atmospheres Company, Hawthorn, CA. These units are operated continuously and the DRI-TRAIN provides primary pressure control for the glove box.

Oxygen and moisture are removed by the DRI-TRAIN. This is accomplished by passing the box gas through alternating layers of molecular sieves and finely divided copper on an alumina matrix (Ridox). The Model MO-120-2 contains two impurity removal beds. Each bed has an oxygen removal capacity of 40 liters and a moisture removal capacity of 9.0 kg at 25°C. Dual beds allow operation of one bed and simultaneous regeneration of a spent bed. Regeneration of an exhausted bed is performed in a 24-hour timed cycle. The spent bed is heated and purged with a 6 v/o H_2 balance He gas mixture to reduce the CuO formed to Cu and to drive off the water produced along with that collected on the molecular sieve. Subsequent cool down, evacuation, and inert gas backfill phases of the cycle ready the bed for reuse.

The Model NI-40 NI-TRAIN also has two beds available for use. The NI-TRAIN operates in parallel with the DRI-TRAIN to remove nitrogen from the glove box atmosphere. Glove box gas is passed through a retort containing 2.3 kg (5 lb) of titanium sponge and operated at temperatures of 900 to 930°C. The nitrogen present in the gas stream reacts with the titanium at this temperature to form a stable nitride. In contrast to the DRI-TRAIN, the spent beds cannot be regenerated to reclaim the Ti and must be replaced. Each bed has 150 liter N_2 capacity.

IMPURITY ANALYZERS

Separate instruments monitor levels of oxygen, moisture, and nitrogen in the glove box atmosphere. Oxygen is continuously monitored with a Vacuum Atmospheres Oxygen Analyzer Model AO-316-H with the High Oxygen Protection System (HOPS) option. A diaphragm pump located inside the glove box continuously draws glove box gas through the solid state trace oxygen analyzer. A micro-fuel cell measures the concentration of oxygen in the gas stream electrochemically. A concentration of 50 ppm O_2 is set as an arbitrary alarm point. Alarm conditions cause isolation of the NI-TRAIN from the glove box since very high O_2 levels may cause an uncontrolled reaction with the Ti sponge at the operating temperature of the bed. This analyzer is routinely calibrated using room air for one calibration point and a He-H_2 gas mixture passed through a deoxo purifier to provide <1 ppm O_2 for a low oxygen calibration point.

A Panametrics Model 700 Moisture Monitor and a matched aluminum oxide cell (this cell is formed by anodizing an aluminum strip to provide a porous oxide layer which is then coated with a thin layer of metal) was obtained from Panametrics of Waltham, MA, is used to continuously monitor the H_2O levels. The cell is installed in the wall of the glove box and is electronically calibrated.

Nitrogen analysis is performed on an individual sample basis using a Vacuum Atmospheres Model AN-1 Nitrogen Analyzer. The analyzer uses a copper column packed with type 5A molecular sieves to separate hydrogen, helium, oxygen, and nitrogen. A high sensitivity thermal conductivity detector is used for analysis. A separate diaphragm pump mounted inside the glove box draws a sample of the glove box atmosphere through this analyzer. The analyzer is calibrated by passing a certified standard gas mixture of H_2 and N_2 in argon and determining the response.

SAFETY

Because of the nature of the materials used, it is imperative that safety systems are integrated into the entire facility. The glove boxes are protected against high and low pressure conditions with Dwyer pressure switches models 1626-1 and 1626-5 which have been calibrated to alarm at set pressures.

A high pressure condition is sensed when a box pressure of $\geq$-0.15 in. H_2O is reached and remains in effect for three seconds. This time limit was established to prevent short-duration pressure excursions which may occur during normal box operations which could then trigger the alarm. A real high pressure condition will result from either an unexpected reaction in the glove box or a large atmospheric leak such as loss of a glove. If a high pressure condition were to occur, the following would take place:

1. Immediately pilot lights on a panel annunciate the alarm condition.

2. After a three-second delay, a signal is relayed to the Central Surveillance System, an audible alarm sounds, and a 150 CFM blower which is ducted separately to each glove box is started. The operating blower forces the duct pressure to be negative with respect to the glove box pressure.
3. After an additional two-second delay (total five seconds), the DRI-TRAIN and NI-TRAIN purification systems are isolated and a 10-cm (4 in.) diameter emergency valve opens and allows the blower to draw on the glove box atmosphere and reduce the pressure and maintain a negative pressure in the box. This prevents the escape of activity from the glove box.
4. The system vacuum pumps are shut down due to a low glove box pressure condition which has now alarmed since the blower reduces the glove box pressure below the low-pressure alarm point.

When the over-pressure condition is alleviated, the system can be manually reset and normal operation of the glove box resumed.

A low-pressure alarm condition can be experienced if the glove box pressure were lowered to $\leq$-2.0 in. H_2O. Possible causes would be inadvertent evacuation of the glove box with a vacuum pump through a furnace well or the malfunction of a vacuum pump used for pressure control and maintained as an integral part of the DRI-TRAIN. In either case, the alarm condition causes the following:

1. Lights on the annunciator panel will immediately light.
2. The vacuum pumps shut down after a five-second delay.
3. A signal is relayed to the Central Surveillance System.
4. An audible alarm will sound.

If the low-pressure condition clears, the external vacuum pump will remain off but can be manually reset and restarted. The vacuum pump mounted in the DRI-TRAIN and used for glove box pressure control will restart when the alarm condition is alleviated so that glove box pressure control may be regained.

An oil bubbler is installed on each of the glove boxes. The bubblers relief pressure was set at ~+4.0 in H_2O in the over-pressure model and ~-4.5 in. H_2O in the under-pressure mode.

ACKNOWLEDGMENTS

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