

**HOT CELLS FOR FUEL CHARACTERISATION AND TESTING
IN RELATION TO DRY STORAGE**

by

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Summary

Dry storage of spent fuel is being considered by many Utilities worldwide as an alternative disposal route to immediate reprocessing. Scottish Nuclear Ltd are currently assessing dry storage as a means of disposal for gas-cooled fuel and have instituted various studies into its drying and corrosion behaviour. As part of this programme a hot cell facility has been set up by AEA Technology for experimental work on spent irradiated fuel. The facility has performed well and according to specification.

1.0 Introduction

The Modular Vault Dry Store (MVDS) concept has been selected by Scottish Nuclear Ltd for storage of spent fuel from the Torness 'A' power station, with another store of the same design planned for the Hunterston 'B' station. The spent fuel will be stored for periods of 50 to 100 years before ultimate disposal by reprocessing or despatch to a repository. The MVDS design is similar to the facility at Fort St. Vrain in the USA built by GEC-Alsthom Engineering Systems and Foster Wheeler Inc. for storage of High Temperature Gas-Cooled Reactor fuel, but differs in detail owing to the special requirements for storage of Civil Advanced Gas-Cooled Reactor (CAGR) fuel. The MVDS will consist of three main items: a fuel receipt cave, fuel storage tubes (FST's) held in concrete vaults built as modules, and a fuel handling machine.

Since CAGR fuel is initially stored under water in the station ponds and is transported in water-filled flasks, it is necessary to provide facilities for drying the fuel elements before transferring them to the fuel storage tubes in the MVDS. Equipment for drying the fuel will therefore be provided in the Flask Unloading and Drying Cave. Since the graphite sleeve and the carbonaceous deposits on CAGR fuel pins have a significant ability to hold water, both in pores and by surface adsorption, a fully engineered system is necessary to ensure that the greater part of the moisture is driven off. The technique to be employed incorporates radio-frequency induction heating of graphite sleeves in conjunction with a hot argon gas stream passed over the fuel pin cluster.

Although the 20Cr/25Ni/Nb stainless steel cladding used for CAGR fuel is normally resistant to corrosion, under certain conditions of irradiation temperature and fast neutron dose it becomes "sensitised", ie, prone to intergranular attack on exposure to moisture and oxygen [1]. Sensitisation is most pronounced in the bottom fuel element of a stringer (element 1 of 8 elements) notably where irradiation temperatures are around 400°C, and may also occur at the ends of pins in adjacent lower elements where similar temperatures prevail. Sensitisation is caused by radiation induced segregation at grain boundaries, probably by the diffusion of interstitial-vacancy complexes [2] generated by fast neutron irradiation. The principal effects are enrichment of the grain boundaries in nickel and silicon, and depletion in chromium. To ensure that no corrosion can occur during storage in the FST's, moisture and oxygen must be maintained at sufficiently low levels in them, which in turn requires prior drying of the fuel to low moisture levels and the use of an inert cover gas. With these considerations in mind, Scottish Nuclear Ltd and AEA Technology, Windscale have instituted a number of development and research programmes, amongst which are investigations at Windscale into the drying conditions required to achieve low retained moisture levels in irradiated fuel, and into the corrosion behaviour of irradiated Element 1 pins during storage. In this paper an account is given of the hot cell facilities at Windscale for corrosion and drying tests to support the Safety Case for the MVDS.

2.0 Experimental Programme

2.1 CORROSION TESTS

Two types of corrosion test were required:

- 1 Exposure of whole fuel elements, ie graphite sleeve plus fuel pin cluster, to conditions similar to those of the FST's but with somewhat more aggressive moisture levels, to obtain lead information on fuel cladding integrity before construction of the MVDS. The main limitation of this type of test is that it can only give a fail/no fail result;
- 2 Exposure of stressed cladding samples to a further range of conditions somewhat more aggressive than those of the FST's, followed by fracture and examination for intergranular attack. With this type of test part-through intergranular cracks can be detected which can be measured and the data extrapolated to provide estimates of corrosion rates over long periods of time. In principle the potential advantage of this technique, which had been adopted in previous studies is that it allows determination of smaller corrosion rates that is possible with the first type of test, and a test matrix could be devised to give information on the time dependence of corrosion and on possible incubation effects.

For the tests on whole fuel elements it was decided that two simulated FST's would be constructed, each capable of holding one Element 1 under inert cover gas and at a temperature representative of MVDS operation. The principal difficulty was that no equipment existed in the hot cell facilities capable of drying an entire CAGR fuel element which had been stored previously under water in the ponds and flask. A particular problem was the graphite sleeve, which was known to hold up to a kilogram of water. It was therefore decided to remove the graphite sleeves from the wet-stored elements and substitute "bottled" sleeves which had never been immersed in water, (bottling facilities exist at the stations in which fuel can be enclosed in sealed stainless steel bottles filled with a dry nitrogen atmosphere before despatch to the pond. The fuel pins of the wet-stored elements were known to hold some water in the carbonaceous deposits on them, but the quantity was less than that held in the sleeve and could be removed by a combination of initial evaporation under the decay heat of the pins, and subsequent cycles of purging and heating with dry gas and chamber evacuations. The fuel elements were then to be held under stagnant atmosphere conditions and a means provided for periodically sampling the cover gas for moisture and fission product gases(ie ^{85}Kr).

For the corrosion tests on cladding samples two containers were to be provided to allow sets of specimens to be exposed to different conditions of temperature, atmosphere composition, strain and gamma radiation field. The specimens would be prepared by removing the fuel pellets from Element 1 fuel pins and then cutting out conventional tensile testpieces using electro-discharge machining. The specimens would be mounted on jigs so that a specified plastic strain could be applied and a force maintained on the specimen throughout the corrosion tests, to simulate the strains and forces experienced by cladding caused by differential thermal contraction after irradiation. A means of breaking the specimens was also needed at the conclusion of the exposure period so that the fracture surfaces could be examined by scanning electron microscopy (SEM) to detect intergranular attack. The tests were to be performed under flowing gas conditions at a low flow rate (approximately 0.5 litre/minute). To avoid possible corrosion initiation effects during pond storage, bottled fuel was specified for the tests.

2.2 DRYING TESTS

The objectives of the drying tests were:

- 1 To establish the temperature, time and flow rate conditions necessary to obtain sufficiently low levels of retained moisture in the fuel elements;
- 2 To measure the activity produced by the spalling of deposit and oxide from the pin cladding surface, since this affects the levels of contamination to be expected at the MVDS Drying Station and its filtration system;
- 3 To determine any effects of drying on subsequent cladding corrosion behaviour;
- 4 To measure the post-test description of moisture.

Drying tests on complete fuel elements were not practical because of the scale of the equipment required, which would have required a large amount of cave space and would also have been very expensive. The drying of graphite and fuel pins were therefore considered as separate topics, with the testing being performed on small samples of graphite and on individual fuel pins. Since graphite is of relatively low activity, drying tests on it did not require heavily shielded hot cell facilities and so are not considered further in this paper. Drying tests were to be performed on Element 1, 4 and 7 pins (the latter being peak rated and peak cladding temperature positions) to obtain a range of deposit and oxidation characteristics; an essential requirement was that the fuel had to be stored under water prior to dismantling and prior to testing. A method for trapping spalled deposit and oxide for post-test activity and weight measurements was required.

3.0 Hot Cell Installation

The experiments are located in the waste packaging and storage caves at a position where minimum interference can occur between the corrosion and drying tests, and other cave operations. This cave also has the advantage of allowing easy transfers of specimens and equipment in and out through the flask port and also through a corridor linking the caves (Figure 1); this is an important consideration since the experiments may need to be moved to allow access to the cave for repair or maintenance work. The height of a fuel element (about 1 m) is such that if the simulated FST's were placed on the cave bench then access with manipulators would be difficult and the experiments would obstruct other cave work, so it was decided to cut the bench and place the simulated FST's and other corrosion and drying test equipment mainly below bench level. The cave bench was therefore cut to give five square holes each 500 x 500 mm, four to accommodate corrosion tests and the fifth for drying equipment. Each hole was fitted with a stainless steel open-topped box reaching down to floor level, which was then welded to the bench around the top edge, so ensuring that containment was maintained. Each box was provided with a removable liner so that any items accidentally dropped into them could be retrieved. A full range of services was installed for each experiment including pipework for controlled atmosphere gas flows, compressed air and vacuum, electrical supplies and instrumentation wiring. To ensure low moisture adsorption characteristics all pipework used for the controlled atmospheres, including flexible hoses, was of stainless steel given a degreasing treatment prior to installation. A services wall plug fitted with heated pipework was also installed for corrosion tests at high humidities.

4.0 Corrosion Testing Equipment

4.1 WHOLE FUEL ELEMENT TESTS

The FST's at the MVDS consist of mild steel tubes of several mm wall thickness, each one capable of holding up to eight fuel elements. The steel is to a low-temperature specification ensuring adequate toughness under cold conditions and is flame-coated on the outside with aluminium to prevent external corrosion. The corrosion experiment containers were manufactured of the same steel and to the same dimensions as an FST, except that they were only long enough to take one fuel element. They were also flame-coated on the outside with aluminium (Figure 2), and the interior was untreated except for shot-blasting (Figure 3) in the same way as an actual FST. The containers were provided with an electric heater in the base so that temperatures similar to those expected in the MVDS could be applied, and two platinum resistance thermometers were mounted on the outside of each container for temperature monitoring and control. Inlet and outlet pipes were provided for passing a continuous gas flow through the container for establishing a controlled atmosphere and for moisture sampling, while a separate pipe was provided for fission gas sampling; all pipe connections were of the "Swagelok" self-sealing quick release type to facilitate remote operation. The container was closed by a steel lid which required sealing to a high standard of leak tightness, since sampling would only be performed every few months or so. Conventional 'O'-ring seals were not suitable for long-term use in a high radiation field and so silver-on-Inconel metal seals were used instead. Metal seals give low leak rates and do not deteriorate with irradiation and elevated temperature service. However, the finish of the contact surfaces must be of high quality and must not be scratched or damaged, thus for remote handling purposes very soft metals such as indium or lead are not suitable. Silver-on-Inconel was chosen as a compromise since silver is moderately soft and gives a good gas-tight seal while being reasonably resistant to handling damage. The seals are manufactured in the form of 'C'-section rings orientated for either external or internal pressure; a pair of each type was used on the containers so that gas-tightness would be maintained under both positive and negative pressure conditions (Figure 3 shows the seal recesses machined in the container flange). Each container was fitted with a pressure indicator reading from -1 to +1 bar gauge.

4.2 Cladding Specimen Tests

The specimen design is a semi-cylindrical pin-loaded tensile test-piece which has been previously used in several investigations into cladding mechanical properties. The specimens of irradiated cladding are prepared by electro-discharge machining after axial cutting of pin sections and removal of the fuel pellets. After machining, the specimens are thoroughly washed in solvent to remove debris and traces of dielectric fluid, and stored in containers with Silica-gel to maintain low moisture levels of about 90 vpm.

Since stress and plastic strain are factors which can influence the corrosion behaviour, it is necessary to have some method for straining the test-piece before testing and maintaining the stress throughout the test. A simple jig device is used for this purpose in which the specimen is placed on two loading pins and a screw turned by a strain application mechanism to obtain the required plastic strain (Figure 4), the strain being measured by a dial indicator. When the strain has been applied, the tensioning device is removed and the specimen remains under stress. The jig and its specimen can then be loaded into the specimen

magazine (Figure 5), which can hold up to 6 specimens. The magazine is placed in a corrosion testing chamber through which a continuous gas flow of controlled composition and moisture content can be passed, so that 6 specimens are simultaneously exposed to the same corrosion testing environment. Metal seals are also used on these chambers. At the conclusion of the exposure period, if a specimen has not already failed it can be broken by applying more strain until it fails, and then the fracture surface can be examined by scanning electron microscopy.

The required test temperature is obtained by placing the corrosion testing chambers into an outer container which has an electric heater in its base and an air mover to circulate the container air and improve temperature uniformity (Figure 6). Four environmental chambers fit into each container. Temperature control and monitoring is by platinum resistance thermometers mounted externally so that they can be replaced remotely in the event of failure without disturbing the experiment.

Gamma radiation fields can increase the rate of corrosion attack by ionisation of the gas and radiolysis of water vapour, so that the radiation field existing within the MVDS must be reproduced. Calculations of radiation dose rates within the MVDS demonstrated that a gamma radiation field of sufficient intensity could be obtained if the cladding specimens could be kept at a distance of 10 mm or less from a stack of short-cooled (100 days) fuel pins. Accordingly a square section holder is located at the centre of the experimental container which is capable of holding a stack of 36 pins at 10 mm from the specimens. The environmental chambers are arranged in a square array at the centres of the four faces of the fuel holder. Short cooled fuel pins in close proximity can produce a significant temperature rise and so compressed air cooling is provided.

4.3 Controlled Atmosphere Supply and Sampling System

Supplies of gas of the correct composition and water vapour content are necessary for filling the whole element containers before closing off valves and leaving the test under stagnant conditions, and also for providing a continuous flow of gas at a low flow rate of about 0.5 litres/minute for the cladding specimen tests. It is also necessary to have sampling facilities for testing for the presence of ^{85}Kr fission product gas, which indicates failed fuel, and also for moisture content. Since the equipment is required to work reliably for long periods, it was designed to be as simple as possible, without complex control systems which could fail. The method of controlling the moisture content of the gas is by dilution of a gas stream saturated with water vapour. Saturation is achieved by passing dry gas from a cylinder supply through two Drechsel bottles in series, each partly filled with ultra high purity water. The Drechsel bottles are immersed in a thermostatically controlled bath the temperature of which determines the water vapour pressure. The saturated gas passes to a mixing chamber in which it is mixed with dry gas in the proportion to obtain the required water vapour concentration. For most corrosion tests the dew point of the emerging gas is normally well below ambient temperature so that no precautions against condensation are needed as it flows to the corrosion tests. However, in some cases high moisture concentrations are needed for accelerated tests and here it is necessary to heat the pipework between the mixing chambers and the in-cave testing equipment. Out of cave, heating is provided by a hot water pump which circulates water around coils surrounding the pipework, and also around a services plug in the cave wall. Inside the cave, a hot air blower is used to maintain the necessary temperature.

Accurate measurement of moisture content is of central importance to the experiments and so a mirror Dew-point hygrometer is used. This type of instrument depends on cooling of a mirror to determine the point at which dew or ice formation occurs, the change in the reflectance of the mirror at the Dew-point being detected automatically, and the Dew-point temperature displayed. A mirror hygrometer thus depends for its operation on a fundamental physical property and produces accurate, reliable Dew-point values. Its main disadvantages are its slow response time and unsuitability for measurement of very low moisture concentrations. These instruments are also expensive and it is not practicable to have a separate one on each gas supply line; it has therefore been arranged to switch in or out of each supply and can measure moisture content on both inlet and exit lines. A pressure transducer is provided adjacent to the mirror unit so that the measured Dew-point can be converted to absolute water vapour concentration. When setting up a gas supply, the gas flow can be passed through an environmental chamber and its moisture content measured on inlet and exit, so that a high degree of confidence in the moisture concentration can be obtained. Lower cost relative humidity measuring instruments are fitted to each gas supply circuit to protect against any accidental moisture excursions; if a set level is exceeded a magnetic valve is closed to shut off the saturated gas supply. The data outputs from all temperature and moisture measuring instruments are continually monitored and recorded by computer. A general view of the gas supply equipment is shown in Figures 7 and 8.

The fission gas sampling equipment is housed within a glovebox together with the hygrometer mirror unit and consists simply of a connection point for a sampling bottle, and a vacuum pump. The sampling hose is connected to one of the whole element containers and the pipe and sampling bottle evacuated. On opening the sampling valve the system is flooded with gas from the container; the procedure can be repeated to obtain several samples, which can be removed for analysis using beta and gamma spectrometry and mass spectrometry. The interior of the glovebox is illustrated in Figure 9. A general view of the in-cave arrangement of the corrosion and drying equipment is shown in Figure 10.

5.0 Drying Test Equipment

Since the drying tests were performed on single fuel pins, the simplest experimental arrangement was a drying tube through which air could be passed at the required temperature and flow rate. Some method of holding and locating an individual pin within a drying tube was needed and also some means of retaining any spalled deposit or oxide. The method used was to provide a holder, called a "slipper", for each pin into which it was placed on arrival at the test cave and immediately transferred to water storage in a six-pin carousel. For the drying tests, an individual inner tube to retain spalled material was provided for each slipper and pin; the inner tube has a port at the bottom end to admit the air flow and also at the top, to allow gas sampling. The air flow was drawn from the compressed air main, dried and filtered, and was then heated in a 6 kW duct heater to temperatures of up to 200°C and passed over the pin at flow rates of up to 700 litres/minute. The exhaust air passed through a sintered stainless steel filter which trapped any particulate material in the air stream for later examination and activity measurement. The main components of the equipment can be seen in Figure 11.

After a drying test on a pin under specified conditions of temperature, flow rate and duration it was necessary to determine the degree of dryness achieved. The drying test was therefore followed by an equilibration test in a separate tube, in which the pin was maintained under

stagnant conditions in dry air for periods of up to 24 hours, at a temperature of 120°C. This treatment drove off adsorbed moisture, which could then be purged from the equilibration tube with a dry air flow and its concentration measured.

In drying tests of this kind it is desirable to measure the moisture concentration as close to the sample as possible, to minimise the effects of adsorption on adjacent surfaces. However, with tests in a cave environment this is not possible since the measuring instrument would quickly degrade when exposed to radiation. The use of a remote sampling system is therefore unavoidable. In the case of the corrosion experiments this is not too serious, since the tests are of long duration and as much time as is necessary can be allowed for gas moisture content to reach equilibrium with the adsorbed moisture. The drying tests, on the other hand, are of short duration and there is not enough time for equilibrium to be established. The solution was to use two sampling lines from the drying tube, one for high level measurement and one for low level, with automatic valve operation to ensure that the low level line only became active when the moisture evolution from the sample had decreased to a preset concentration. The requirement for rapid response also precluded the use of a mirror hygrometer, and the instruments selected for this application depend on the change in capacitance of a moisture sensing device. In addition to shorter response time, measurement to very low moisture contents is possible down to about 0.1 vpm (a Dew-point of -80°C), although the accuracy is not as high as a mirror hygrometer. The output data from these instruments was monitored and recorded by computer during each test; an example of a test record obtained is illustrated in Figure 12 and shows the initial rapid rise in moisture content as water is driven off, followed by a rapid decline, in this example there is also some secondary moisture evolution.

6.0 Conclusion

A novel facility for studying the corrosion and drying characteristics of irradiated CAGR fuel to support the Safety Case for the MVDS has been set up at AEA Technology, Windscale. The experimental facility is the first of its scale to be designed, manufactured and installed in accordance with the AEA Quality Management system and the equipment and instrumentation has performed according to specification.

7.0 References

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- 2 OKAMOTO P R, REHN L E and AVERBACK R S. Solute redistribution processes in ion bombarded alloys. 1982. Journal of Nuclear Materials, vols 108 and 109, pp 310-330.

8.0 Acknowledgement

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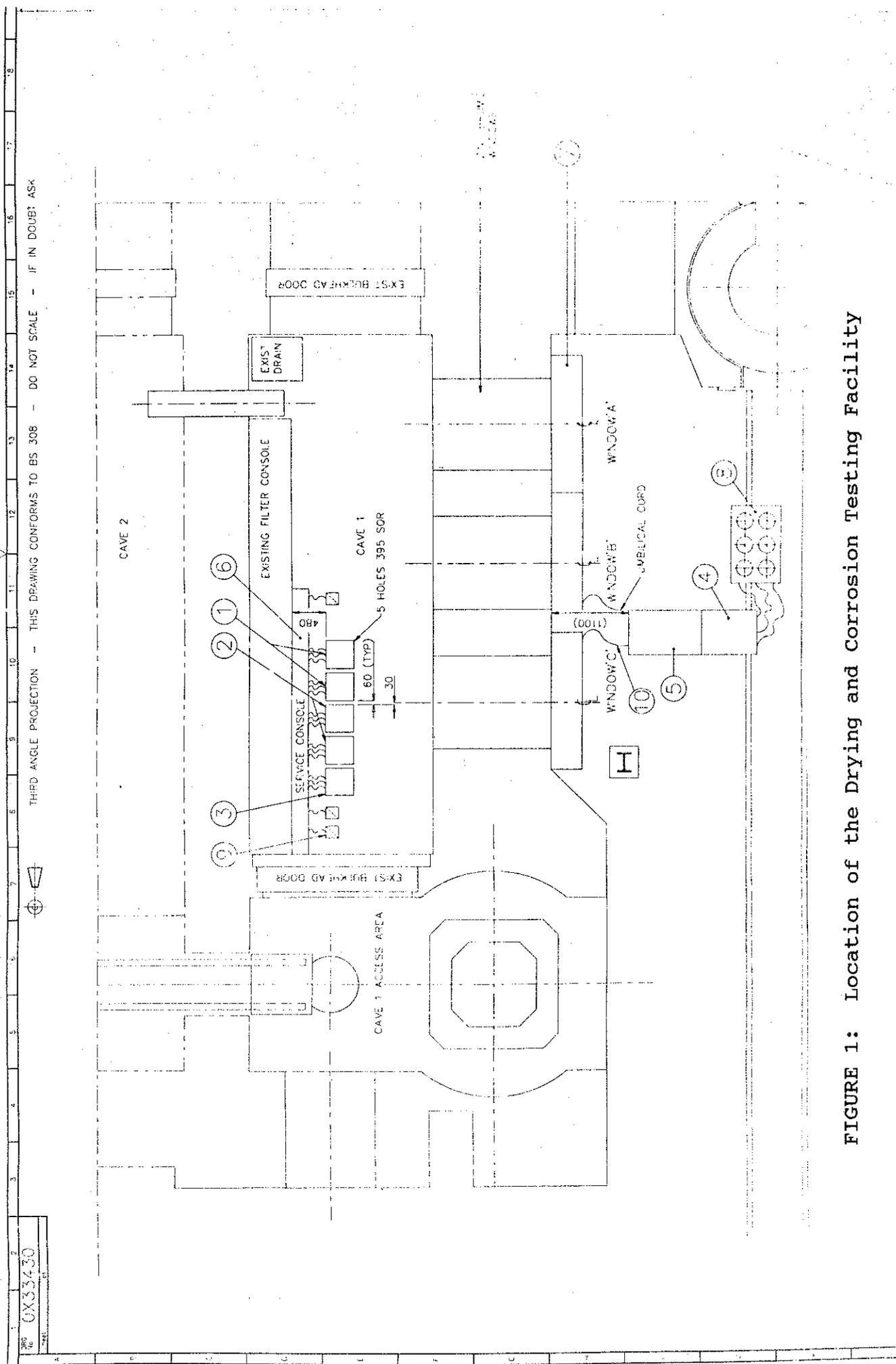


FIGURE 1: Location of the Drying and Corrosion Testing Facility

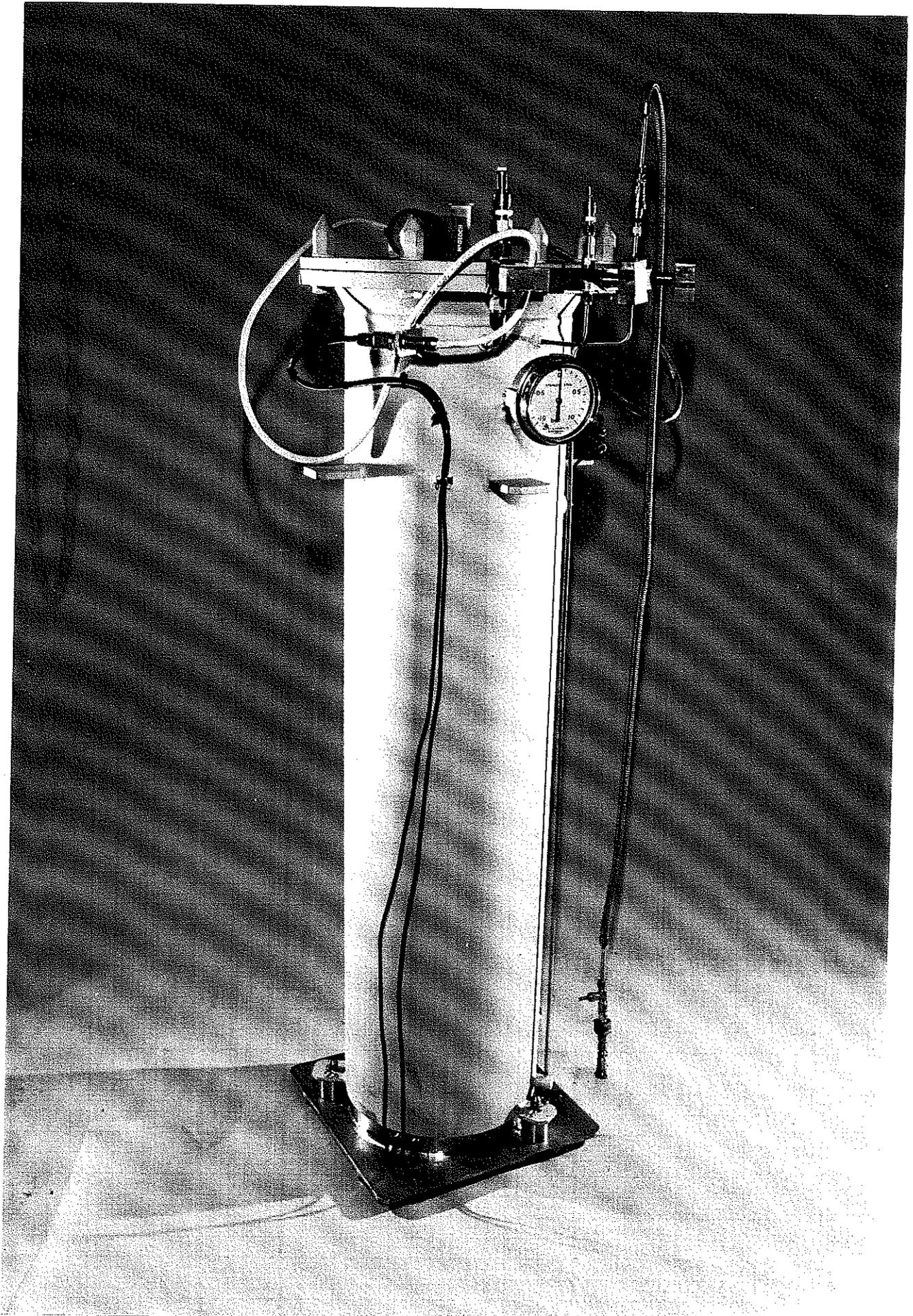


FIGURE 2: Whole element test container

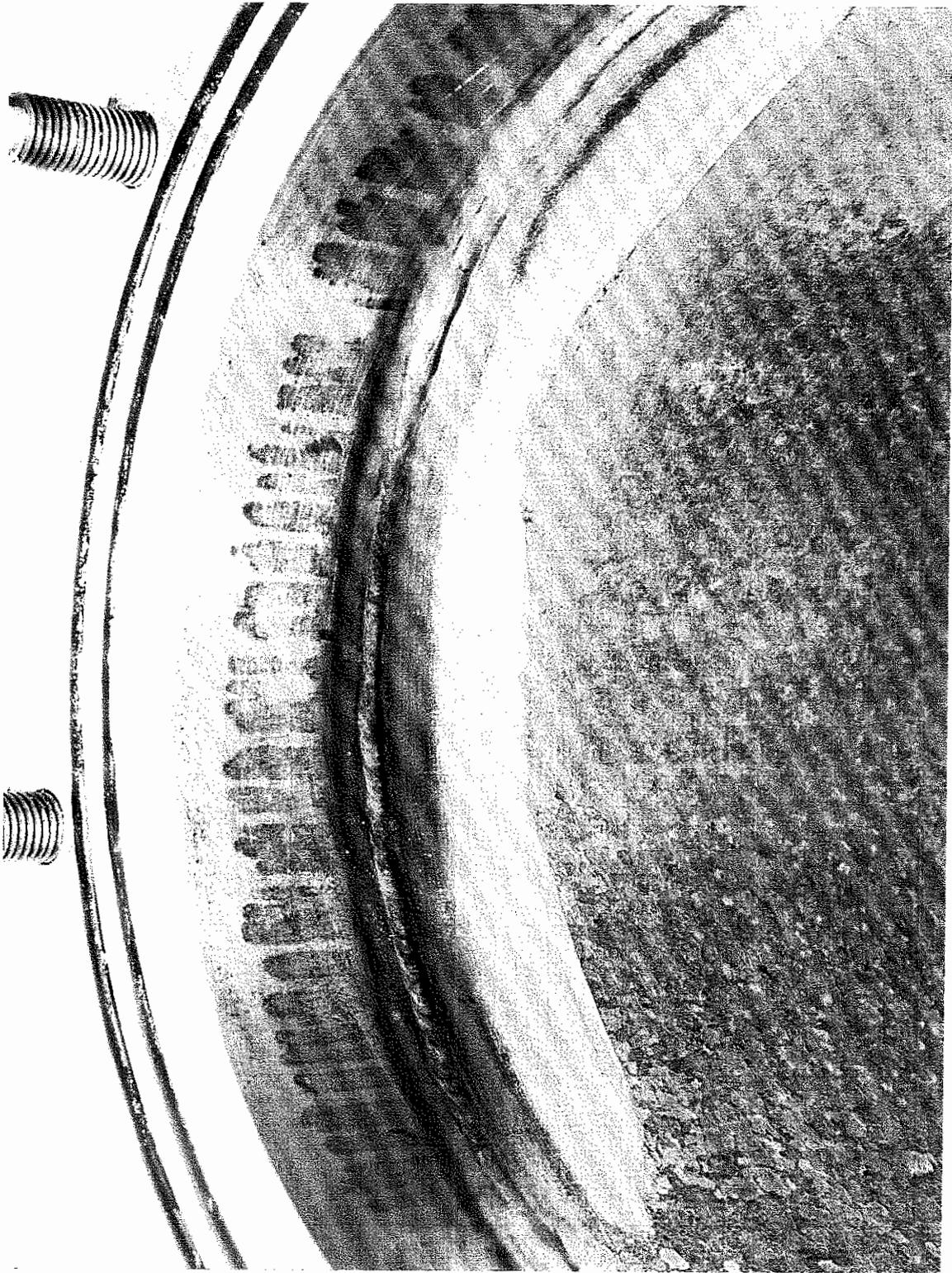


FIGURE 3: Interior view of whole element test container

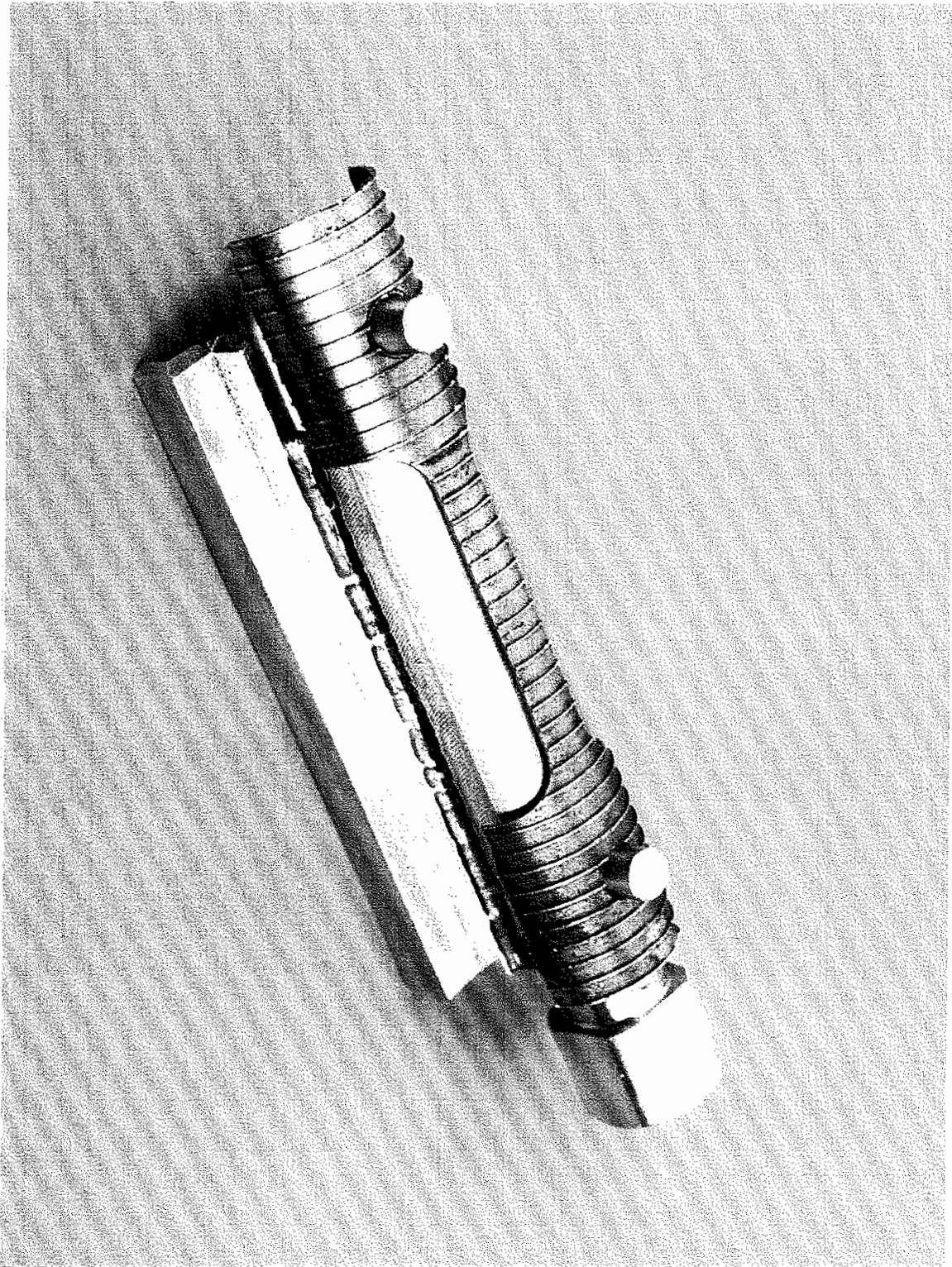


FIGURE 4: Cladding specimen in place on a jig

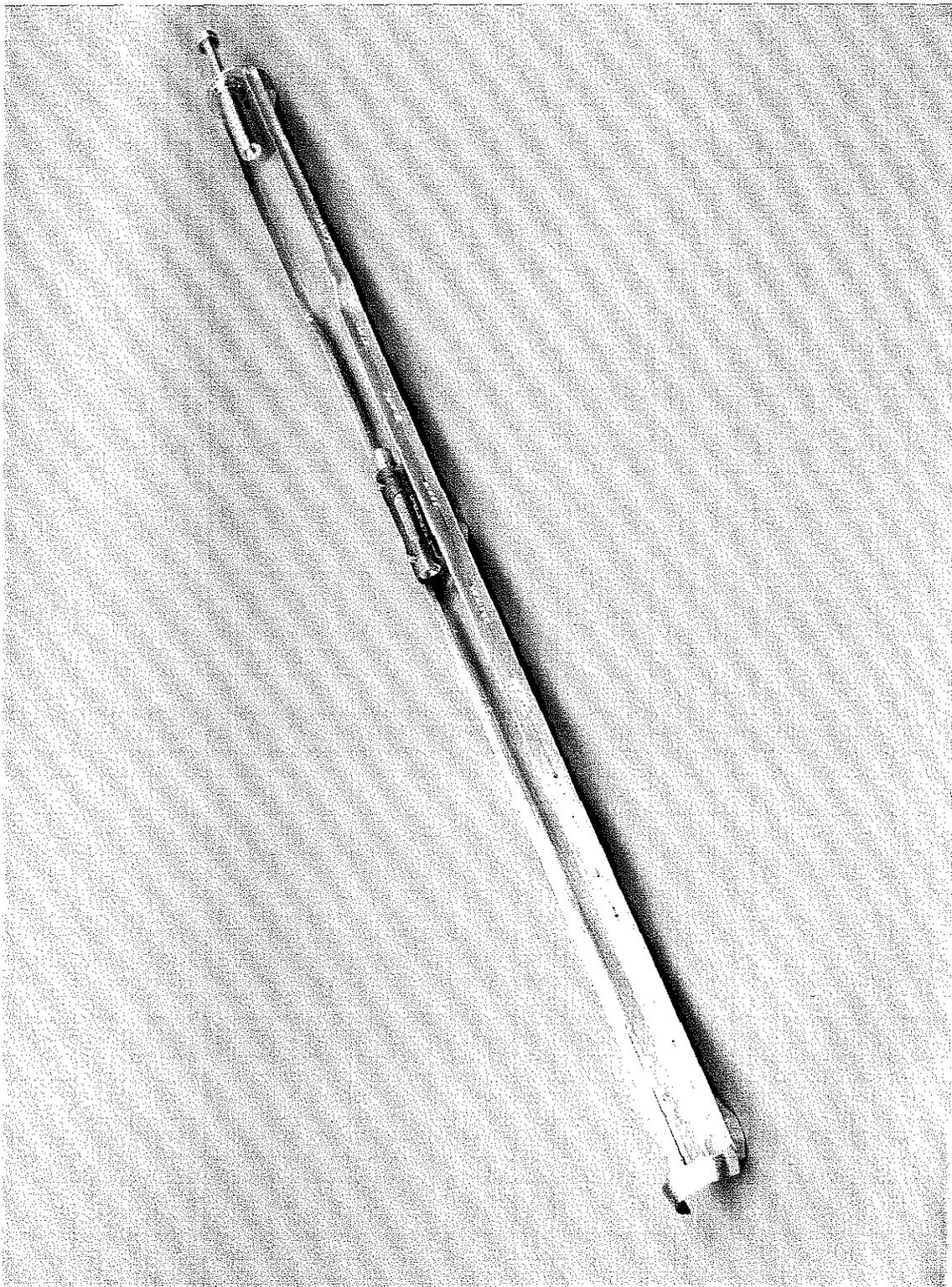


FIGURE 5: Specimen magazine with a cladding specimen and jig

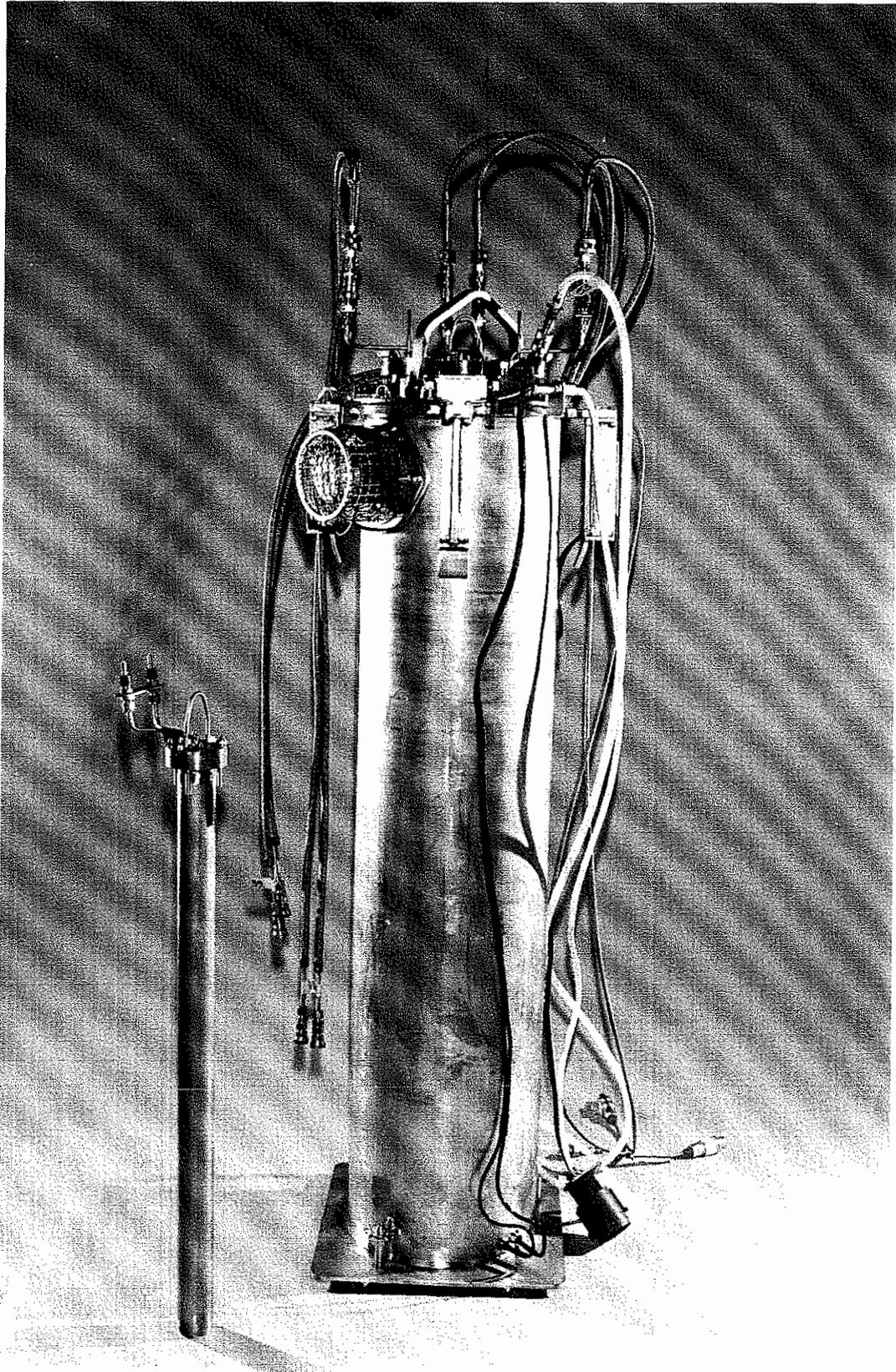


FIGURE 6: Container for cladding specimen tests

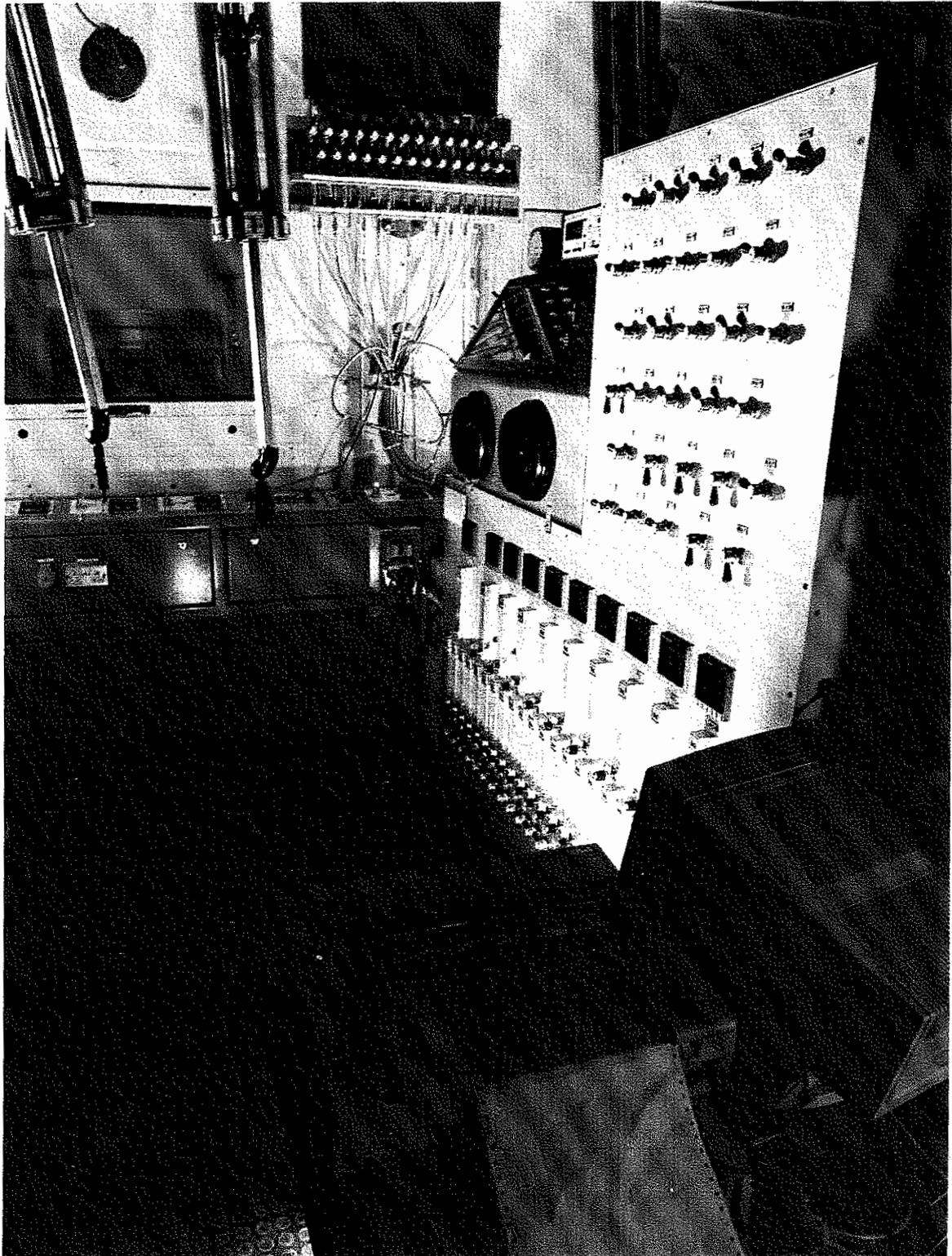


FIGURE 7: Gas supply equipment, view on operating console



FIGURE 8: Rear view of gas supply equipment and drying control console

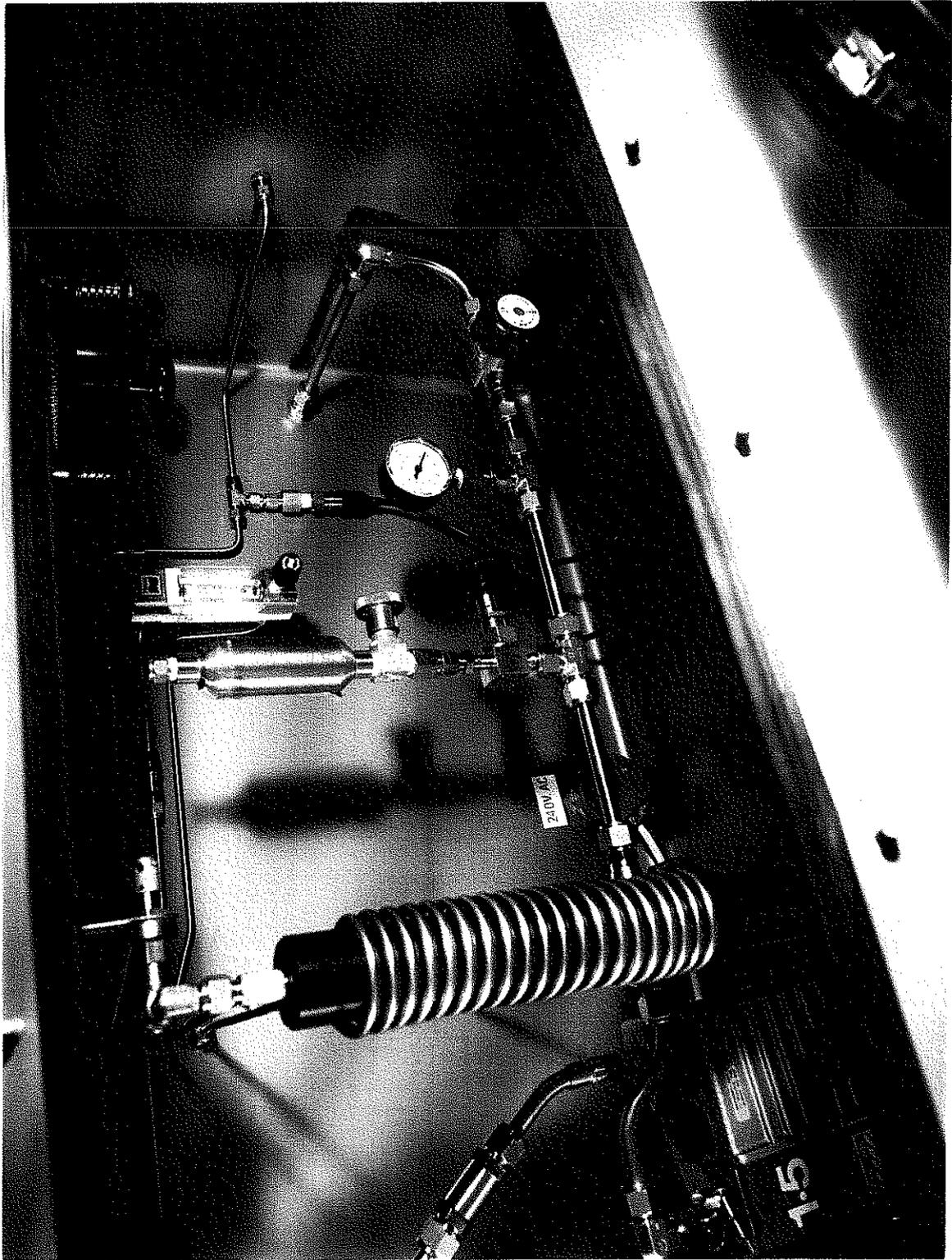


FIGURE 9: Interior of sampling glovebox

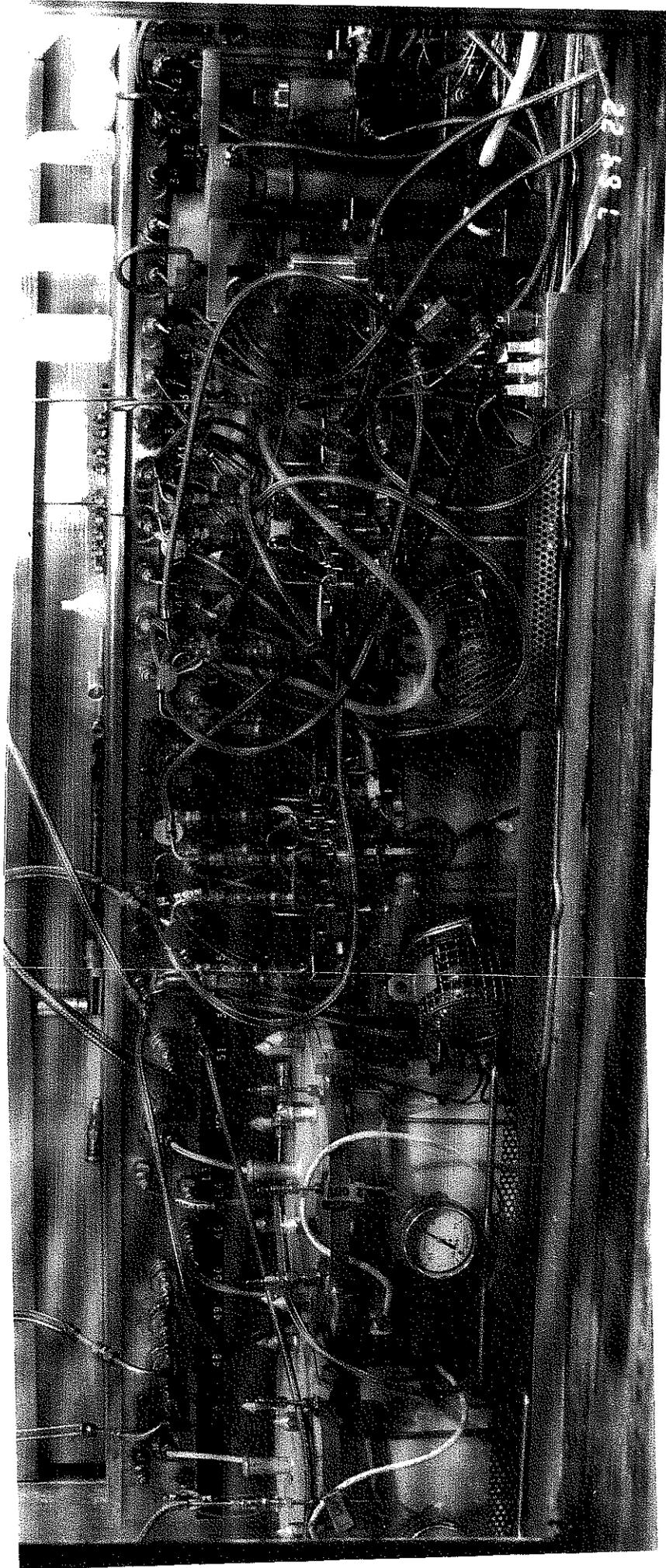


FIGURE 10: General view of in-cave corrosion and drying equipment

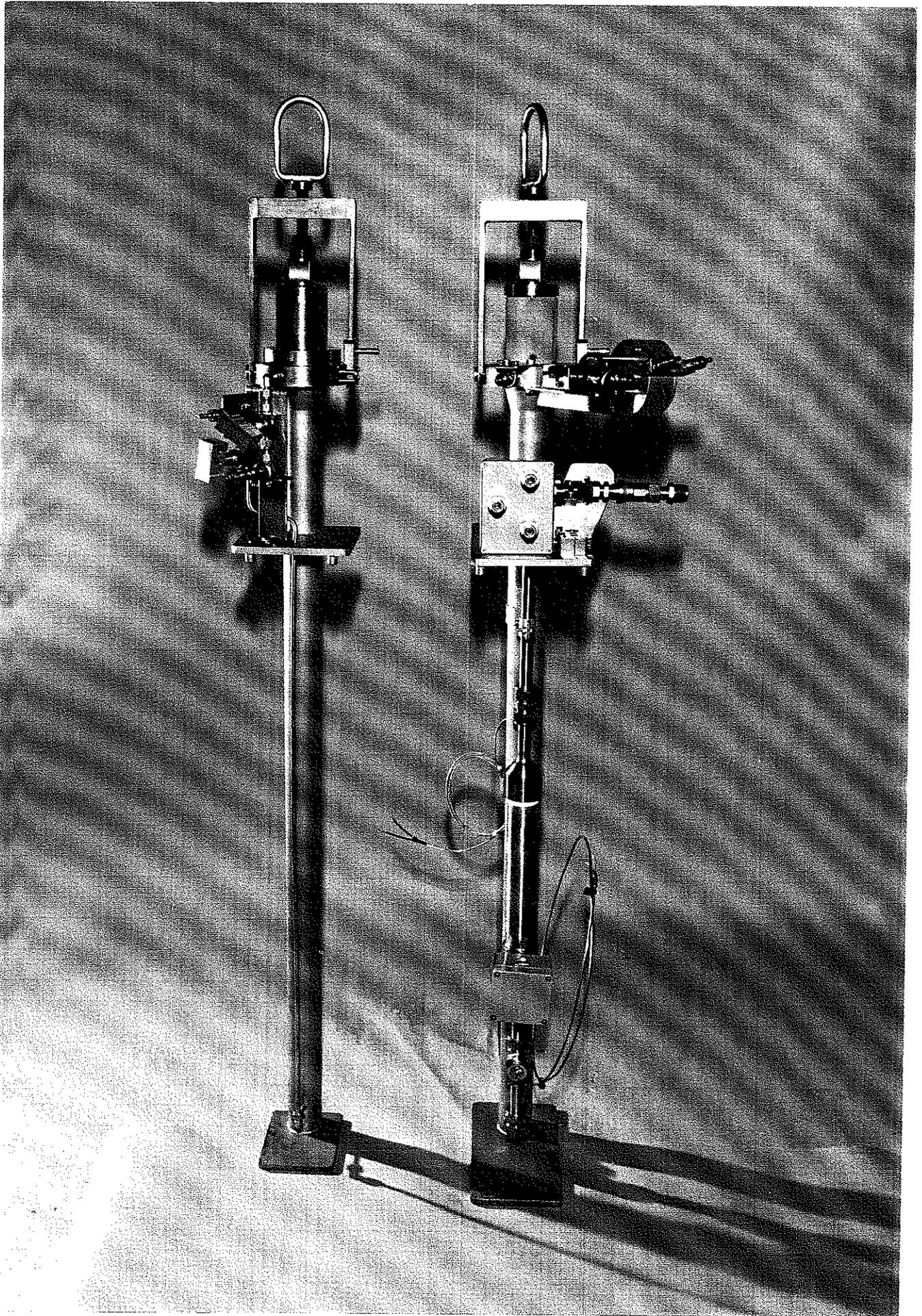


FIGURE 11: Drying and equilibration tubes

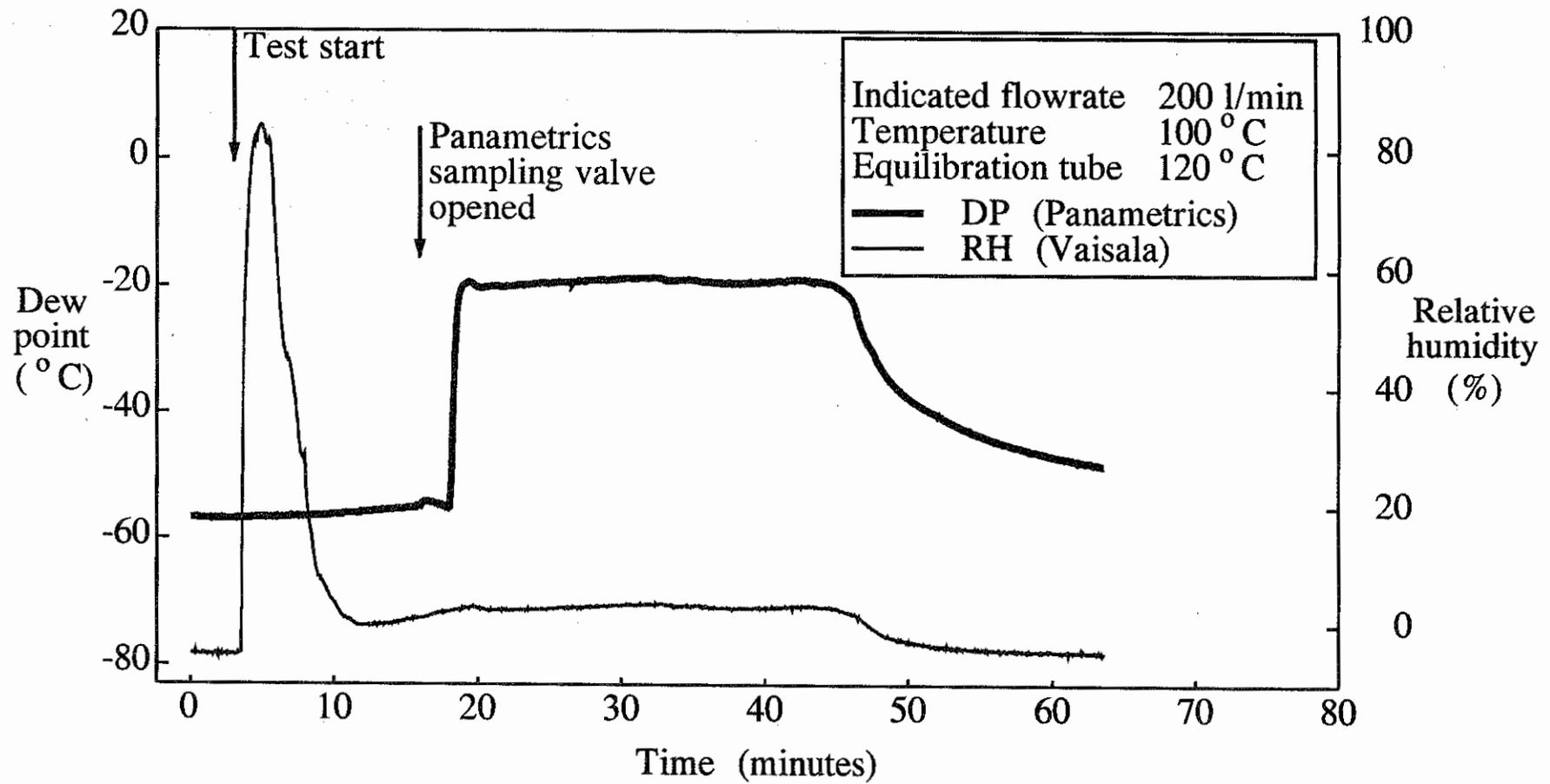


Figure 12: Typical Drying Test Record.