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A HIGH PURITY INERT ATMOSPHERE CELL SUITE FOR RESEARCH PURPOSES

- DESIGN AND OPERATING EXPERIENCE

by

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SUMMARY

The need to prepare and examine irradiated nuclear fuel samples which may contain alkali metal and other oxygen and moisture sensitive materials, has prompted the development of a small specialised hot-cell suite where high atmosphere integrity is the prime consideration. The facility achieves a nitrogen atmosphere with oxygen and moisture levels each below 50 v.p.m. This is two orders of magnitude (x100) better than the inert atmospheres maintained in the normal cells.

Provided certain precautions are taken, metallographic mounting, preparation, microscope examination and sample transfer to remote analytical instruments can be carried out under adequate α-containment and $\beta\gamma$-shielding without significant degradation of atmosphere purity and consequent decomposition of the materials of interest. In the initial months of operation there has been considerable "down-time" due to leaks and atmosphere control failures, but these problems have been overcome and experience suggests that with some design modification and due attention to good housekeeping, such high integrity facilities can be routinely operated without difficulties.

1. INTRODUCTION AND OBJECTIVES

Over the past ten years there has been increasing interest in chemical aspects of fast-reactor fuel pin behaviour such as the role of fission product caesium in clad attack, and the nature of fuel/coolant interactions. Since the materials involved are very reactive alkali metals and their unstable compounds, highly controlled atmospheres are required during post-irradiation examination and analytical stages if meaningful results are to be obtained.

A special high-integrity hot cell suite has been developed in the active handling laboratory of Metallurgy Division, Harwell, with the following objectives:

1.1 To receive sections of pins, cut off in the normal "inert" cell line with special precautions to minimise fuel decomposition.

1.2 To provide ceramographic preparation and examination facilities under water and oxygen free conditions.

1.3 To provide high-integrity storage of reactive samples.

1.4 To provide a means of transferring prepared samples from the suite to analytical facilities such as Scanning Electron Microscopes, Electron Probe Microanalysers and X-ray Diffractometers without atmospheric contamination.

2. SPECIFICATION

2.1 Oxygen and moisture levels should each be below 50 v.p.m. in flowing nitrogen at a depression of -3.0 cms water gauge.

2.2 The α-containment should allow handling of plutonium ceramic fuels with facility for reducing final smear contamination of samples to below 10 c.p.s. alpha, before transfer to other facilities. Sufficient shielding should be provided to handle small fuel sections up to 25 curies $\beta\gamma$ of mixed fission products.

2.3 Provision shall be made for posting in and out without serious degradation of the atmosphere, which should be monitored continuously during operations, with an immediate warning if a preset oxygen level is exceeded.

3. DESIGN AND CONSTRUCTION

3.1 Cell suite (Fig 1)

This consists of three interconnected steel α-box units each approximately 0.75 m square by 0.5 m height and volume 0.3 m^3, with side entry tong manipulators and posting ports. 100 mm lead shielding surrounds the whole suite including bottom and top surfaces. Shielding windows are set in the latter. Lighting is by four 60 watt strip-filament lamps fitted between the shielding and the Perspex top of each box.

Cells 1 and 2 are for preparation or storage and cell 3 is fitted with a remote operated horizontal microscope (1). The in-cell part of the latter is specially designed to pass through a standard 9 inch (23 cms) port for ease of maintenance.

Standard 9 inch bag-posting ports with shielded O-ring sealed bungs are fitted to each preparation cell. A special port is fitted to the microscope cell to mate and seal with the high-integrity transfer flask described in 3.3 below.

As originally designed, conventional tong manipulators working through oiled lead ball units were sealed with custom made double skin PVC gaiters; but, as described in 4. below, some of the ports have now been fitted with single skin bellows-type gaiters with a reinforced mouth.

3.2 Atmosphere Control System (Fig 2)

The suite is supplied by a separately metered nitrogen line direct from the storage tank, without any interconnection with the rest of the building cell-line system. Each box unit is supplied independently by a once-through flow at 20 l min^{-1}, and extracted by air ejectors through filters to the main building extract plenum. Cell pressure is controlled by North regulators (2) fitted across the inlet and extract line to each cell. Pressure surges due to sudden tong movement are damped by conventional oil reservoir chambers.

3.3 High-integrity Transfer Flask (Fig 3)

This is designed to transfer mounted and prepared microsections from the cell suite into other facilities such as an Electron Probe Microanalyser without atmospheric contamination. It consists of a 220 x 150 x 150 mm shielding block containing a 40 mm square-section chamber. The chamber can be sealed by a full-way lever valve with a single 90° action. A push rod fitted with a sample holder passes through a seal in the back of the chamber, and can reach through the opened valve, out of the flask and into the microscope cell.

The flask is carried on a trolley fitted with a rack and pinion for height adjustment. When a specimen is to be loaded, the trolley is secured to positioning lugs on the side of the cell. A tunnel bag is fitted between flask and cell and purged with argon through a hole which is then seal welded. The inner (active) bag is then removed and the flask moved horizontally on slide bars so that its snout with O-ring seal plugs into the mating port entry.

3.4 Atmosphere Monitoring

A gas switching manifold allows selection of any cell or the inlet nitrogen supply for monitoring purity. The sample stream at 130 ml min^{-1} is taken from the filtered flow leaving each cell. It then passes in parallel through a Hersch-cell oxygen meter (3) and a phosphoric acid moisture meter before return to the main extract line. A small diaphragm pump is used to provide adequate sample pressure at the meter inlet to minimise contamination by air in-leakage.

Both the oxygen and moisture meters operate over the ranges 0-10, 0-100 and 0-1000 v.p.m. with a sensitivity of 1 v.p.m. in the 0-100 range. The meter outputs are fed to a continuous chart recorder with an alarm on the oxygen channel which can be set to any desired level in the operating range. Once triggered the alarm must be manually reset, and can also be connected to cut power to in-cell equipment.

4. COMMISSIONING

The preparation cells were equipped with grinding and polishing lap machines and ultrasonic bath cleaners. Cell 1 was appointed for initial receipt and the dirtier grinding operations, whence the sample would be passed into Cell 2, via a 10 cms diameter air-lock transfer tunnel, for polishing and cleaning operations. Similar transfer would be finally made into the microscope cell 3 for examination and removal to other facilities. Such progression was to ensure that the best atmosphere can be maintained in Cell 3 where samples may have to remain exposed for comparatively long periods.

4.1 Leak testing

The alpha-boxes were initially tested to the code of practice AECP(R)5 Method 'B' which specifies a maximum leak rate of 0.05% box volume per hour at -4 inches (10 cms water gauge. After the first fitting and sealing of the tong-shafts and microscope assembly, results were very disappointing and no cell showed better than 1000 v.p.m. (0.1%) oxygen. Exhaustive checks and refitting of many connections and seals indicated no design faults, the high impurity generally being due to the combined effect of many small leaks and some incorrect connections. These problems illustrate that very careful assembly and attention to detail is essential if high integrity is to be achieved.

After the refitting, impurity levels were reduced to a few hundred v.p.m. but obstinately resisted further reduction, particularly in the microscope cell. Further tests revealed that leaks were being created in tong-shaft gaiters during fitting and careless use involving stretching and rubbing against sharp edges. It was also found that some of the internal microscope seals were imperfect.

4.1.1 Gaiters

The frequent leak testing of gaiter assemblies has led to the development of sensitive detection procedures and improved replacement routines. In a "normal" cell the standard procedure, if leaks are detected but not located, is simply to change each gaiter in turn until an improvement results, or even to save time by replacing all gaiters at once by new ones. This involves removal, at each change, of the entire lead ball and shield assembly. To minimise this heavy and dirty task with the high-integrity suite, and to introduce a greater degree of sophistication, the leaking assembly is first located by the following method.

The tong-shaft is removed and clean argon is passed at high flow (20 $l\ min^{-1}$) right down into the "toe" of the gaiter via a tube inserted through the hole in the ball unit, vacated by the tong-shaft. The cell concerned is monitored continuously, and a leaking gaiter usually reveals itself by a marked fall on the oxygen recorder trace within a few minutes. The ball unit is then removed and a bung inserted into the gaiter mouth to stop the leak until the gaiter is changed.

To effect a change with minimum deterioration of the cell atmosphere, the bung is removed, the new gaiter assembly is inserted into the old one and the space between argon purged. The new gaiter is then fitted and the old one withdrawn into the cell for disposal via a trash posting.

4.2 Assessment

Systematic application of the above procedures has resulted in achievement of impurity levels below 50 v.p.m. It has also revealed that the use of double-skin gaiters, which were expected to improve integrity, may often have the opposite effect. Cuts and pinholes caused by sharp objects usually penetrate both skins because they are in close contact. However, the leak may be intermittent and hard to detect. This is because the two holes move apart and become temporarily sealed by adjacent undamaged skin.

In addition, the larger volume of double-skin gaiters causes greater pressure fluctuations during tong movement, and consequently higher flow through any leaks present. A return to the use of lower volume single bellows-type gaiters has improved matters. Although pinholes and tears may be more frequent, the leaks caused are less severe and easier to detect.

5. OPERATING PROCEDURES

5.1 Cutting

3 mm thick sections of fuel pin, containing the feature of interest, are cut off in a normal cell, whose atmosphere is optimised to $< 0.5\%$ oxygen (5000 v.p.m.) by increased nitrogen flow for several hours without postings. The cutting is done dry and as quickly as possible. The section is immediately placed in a sealed aluminium can filled with pure dry argon and containing a slug of sodium metal as a "getter".

5.2 Posting in

During posting into the high integrity suite, the outer clean bag is purged of air with pure argon before seal welding. After posting and re-closing the bung door, the now double bag is cut open. The oxygen level typically rises to ~ 500 v.p.m. during posting, so the sample can is not opened until the cell has recovered to < 100 v.p.m., usually $\frac{1}{2}$ - 1 hour. Moisture level changes are insignificant (< 10 v.p. provided water retentive materials such as tissues are not posted in.

5.3 Ceramographic preparation

All liquids used must be water free and inert to alkali metals. They are always posted-in in well filled containers to minimise trapped air volumes. Light grade liquid paraffin, checked for moisture by metallic sodium, is used as a grinding lubricant with silicon carbide papers. Water free diamond pastes (4) are used with nylon laps for polishing, and cleaning is by liquid paraffin in ultra-sonic baths. Final rinsing to remove dirty paraffin and produce a stain free surface is done with tri-chloro-ethane (SS21) dried over calcium chloride for at least 1 week before use.

Specimens are recanned with sodium getters during break periods in operations, and immediately the oxygen level alarm (> 90 v.p.m.) sounds while operating. The oxygen and moisture level of each cell is checked before cell-to-cell transfer is permitted. Before examination, the microscope is switched on for a period to clear any water vapour released by heating from the illumination train.

6. EXPERIMENTAL RESULTS

The suitability of atmospheres of < 100 v.p.m. purity was assessed by exposing freshly cut sodium surfaces. These remained bright and shiny for up to 2 hours. Previous experience gained in sodium handling operations has indicated that surprisingly high oxygen levels may be tolerated provided moisture levels are very low. This has been confirmed in the present work when moisture levels were often below 10 v.p.m. with oxygen levels above 50 v.p.m.

Polished fast-reactor fuel sections containing sodium and sodium/fuel interaction products have been repeatedly examined and photographed without visible decomposition. In some cases, sections were deliberately left on the microscope overnight at oxygen levels ~ 100 v.p.m. The slight reaction which takes place produces a self-etching effect and can thereby positively assist the distinction of phases. Even after long storage times in the suite (> 2 months) canned sections were not degraded beyond recovery by re-polishing.

Prepared sections have been transferred to an Electron Probe Micro-analyser, modified to accept the special flask, without significant degradation. Even transfer to unmodified and air filled facilities, including a Scanning Electron Microscope and an X-ray Diffractometer, proved possible if the sample was protected by a hydrocarbon spray coating or enclosure in thin plastic film.(5)

7. CONCLUSIONS

With careful housekeeping and rigorous precautions during posting, storage and operations, reactive samples have been prepared and examined with satisfactory results, indicating very little decomposition.

In the light of experience, the suite would benefit from higher nitrogen flow to give faster recovery from atmospheric contamination.

The rather small size of the cells coupled with the "top only" windows has made work difficult, especially for short operators.

All operations were bedevilled by leakage from gaiter failures. These could be reduced by the use of a small power manipulator working through the roof of each cell and capable of the high-stress operations such as door closing, equipment moving and maintenance. However such improvements would be a costly addition to an already expensive facility.

8. ACKNOWLEDGMENTS

The author is merely a user of the facility described, which was designed against a requirement for which there was little practical experience. He wishes to record his appreciation of the initial design and assembly work carried out by Engineering Projects Division from a specification of Mr. F.T. Ewart of Applied Chemistry Division; also the design detailing by Norris Brothers Ltd. and the excellent horizontal microscope made by Applied Optics Ltd.

He is particularly grateful for the painstaking operations and leak-hunting carried out by Mr. A. Rollo and other staff of the Active Handling Laboratory, Building 393.6.

9. REFERENCES

1. Microscope by Applied Optics(Coulsdon) Ltd.
2. North regulators by Moores (Wallisdown) Ltd.
3. Hersch Oxygen Meter Mk. II by Engelhard Industries.
4. Hyprez LHT Grade Diamond Pastes by Engis Ltd.
5. Domestic "Cling Film" by Stuart Edgar Ltd.

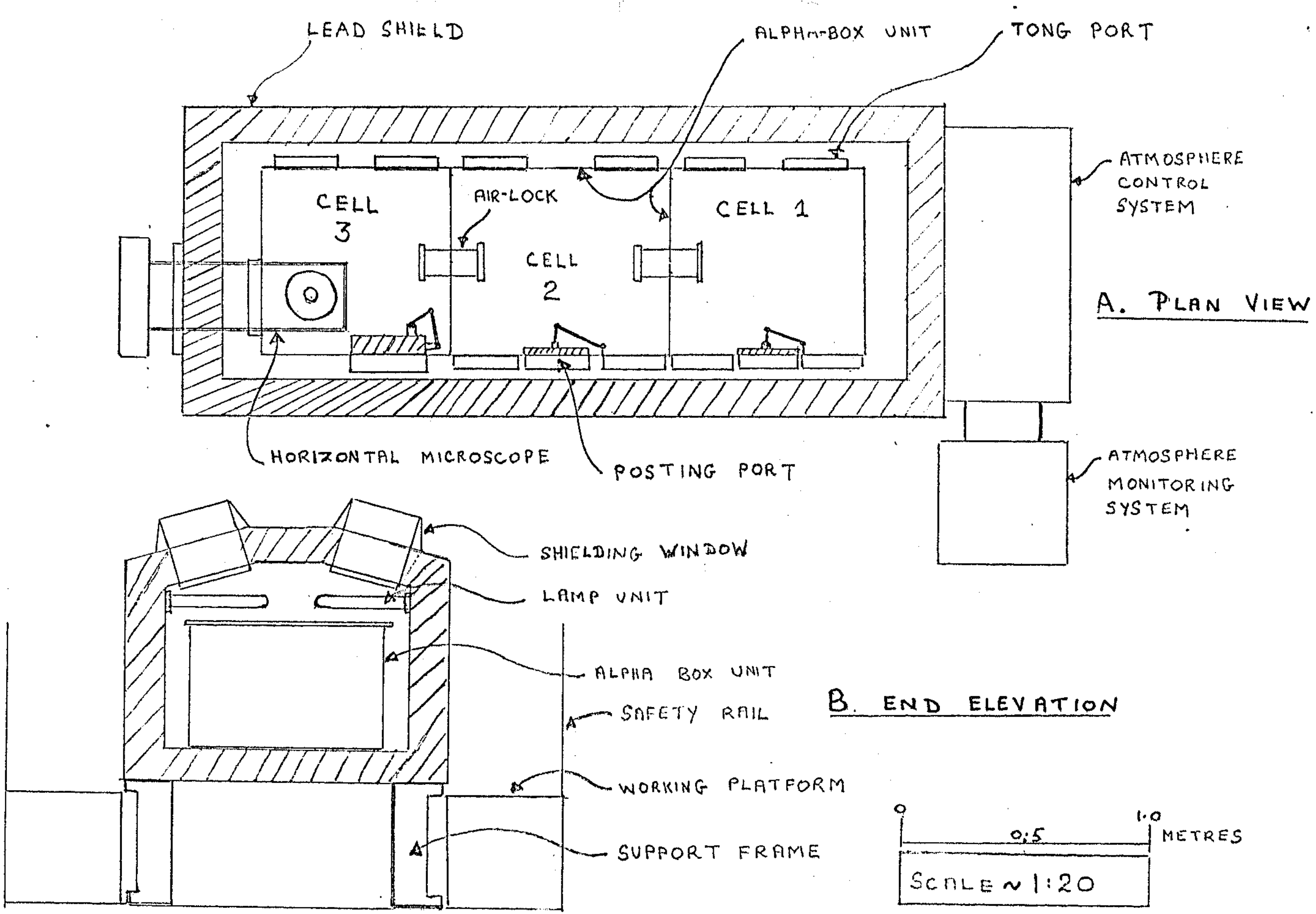

FIGURE 1 – LAYOUT DIAGRAM OF HIGH-INTEGRITY CELL SUITE

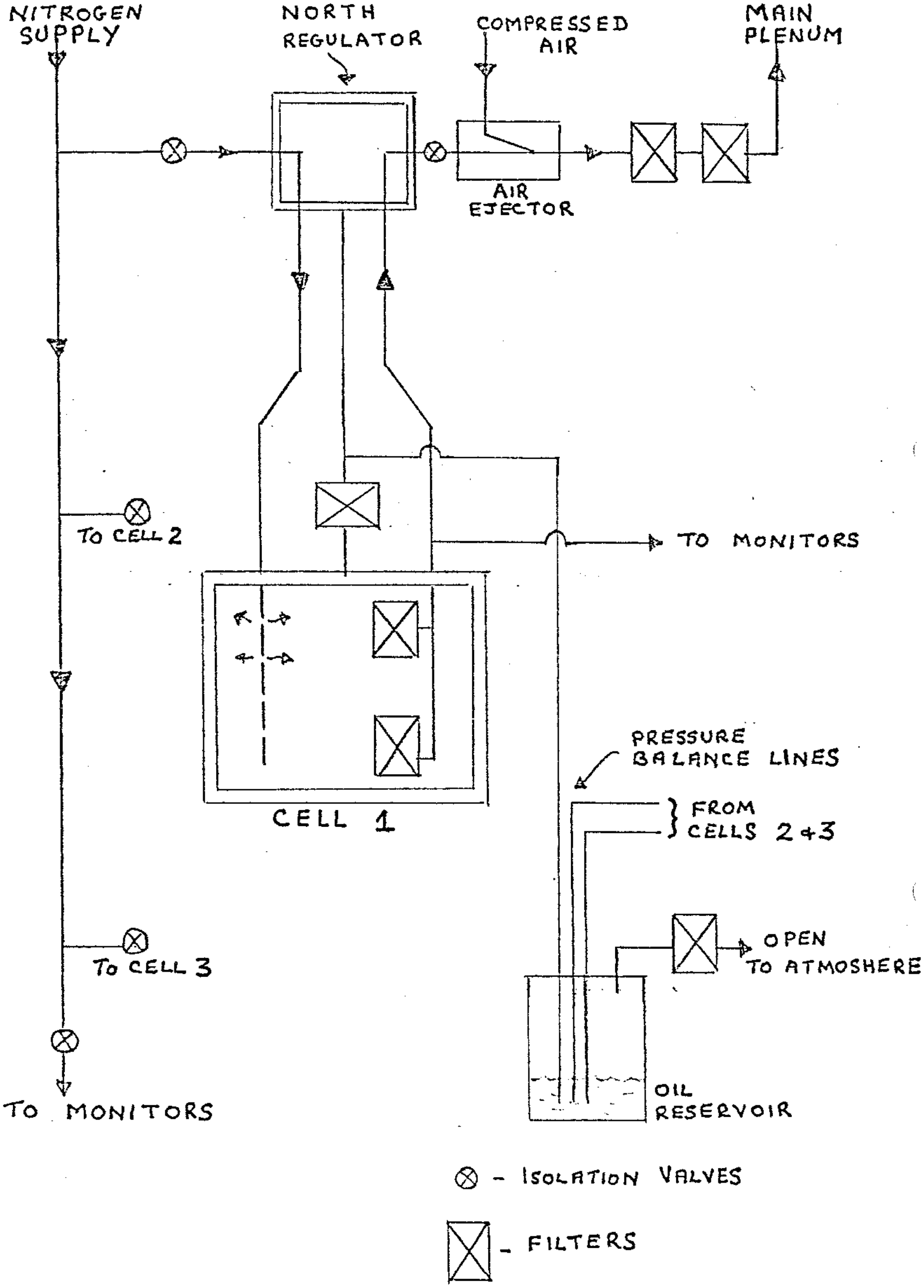

FIGURE 2 - ATMOSPHERE CONTROL SYSTEM

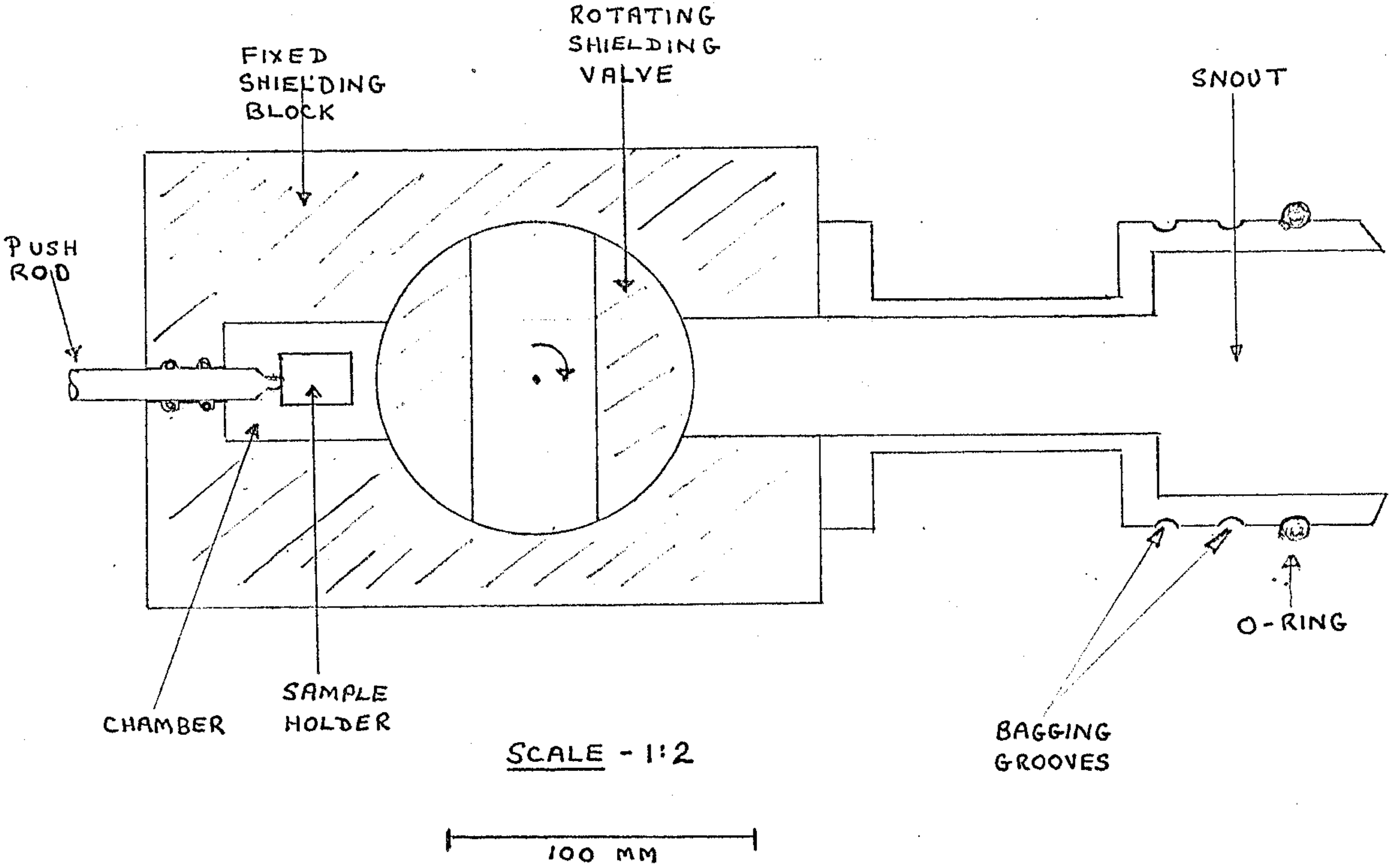

FIGURE 3 - HIGH INTEGRITY TRANSFER FLASK (PLAN VIEW)