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EUROPEAN ATOMIC ENERGY COMMUNITY - EURATOM

PROCESS CELL AND EQUIPMENT FOR
TRANSPLUTONIUM ISOLATION
AT KARLSRUHE

by

W. MÜLLER

1967



Joint Nuclear Research Center
Karlsruhe Establishment - Germany
Transplutonium Elements Program

Paper presented at the
International Transplutonium Elements Symposium
Oak Ridge National Laboratory,
Oak Ridge, Tenn., USA, November 8-10, 1966

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SUMMARY

At the European Transuranium Institute shielded cells have been prepared for the processing of transplutonium targets. The equipment designed for this processing is described.

1. Introduction (*)

The European Transuranium Institute at Karlsruhe is one of 4 installations which form Euratom's Joint Research Center. Although today the main interest of this institute involves research with and technology of plutonium and its compounds, an increasing part of the future activities will be concerned with transplutonium elements.

In a first stage of the Euratom transplutonium program, KOOI processed one americium slug which had been made available by the AEC after irradiation in the MTR reactor at Idaho Falls. The results of this processing which took place last year, have been published.^{1, 2}

In the meantime, 3 capsules prepared at the Livermore Laboratory, have been irradiated in the reactor BR II at Mol/Belgium. The three targets contained initially 3.4 g ^{241}Am in form of 7 % americium oxide-aluminum pellets. They have been exposed to an integrated flux of $1.7 \times 10^{22} \text{ n/cm}^2$, the average fluxes in the different positions of the reactor varying from 1.5 to $9.0 \times 10^{14} \text{ n/cm}^2 \text{ sec}$. After a cooling period of approximately 5 months, the capsules have recently been shipped to Karlsruhe. We intend to process these targets at Karlsruhe and to reirradiate a part of the Am-Cm fraction.

A new set of targets is being prepared. Aluminum cermets containing a total of 25 g ^{241}Am as oxide have been pressed. The pellets will be encapsulated with aluminum and - after welding - will be irradiated at Mol.

I want to describe briefly the process cells available and the equipment to be used for the heavy element processing at Karlsruhe.

(*) Manuscript received on November 11, 1966.

2. Process Cell

2.1 Hot Cell Facilities at the European Transuranium Institute

The institute building contains a wing (B) with 2 different rows of shielded cells, one for metallographic and physical studies of irradiated fuel elements, the other one for analytical and transplutonium chemistry (Fig. 1). A central limited access area separates the two lines with their corresponding operating areas from each other (Fig. 2). Besides the offices and the workshop, there are cold laboratories for the preparation of cold reagents and equipment. The analytical and α -laboratories are in wing A.

2.2 Description of the Chemistry Cells

The 6 chemistry cells arranged in line contain each a stainless steel plated, α -tight containment box (Fig. 3). The surface of the floor is 4 m^2 , the volume of the cubicle is about 14 m^3 . The γ -shielding is provided by 90 cm concrete with a density of 3.5 g/cm^3 (Fig. 4). The lead glass windows - they contain each an oil layer which is 104 mm thick - are equivalent to the concrete for γ -attenuation. The optical contact between the different layers of heavy glass (density 3.5 and 4.2 g/ccm) is made by oil; the optical transmittance for sodium light is almost 40 %. A stabilized special glass covers the inside of the viewing window. No special neutron shielding is foreseen for the moment. Except the ports for the manipulators and the periscope, there is no direct connection between the containment box and the operating area in front of the cells. The whole cubicle is painted with stripping coat.

2.3 Transfer of Material

All cells are interconnected by a magnetically operated conveyor (350x200x300 mm) which is accessible through a glove box at one end of the cell line (Fig. 5). Highly active material has to be introduced or disposed of through a rotary transfer port in the rear concrete plug. Small samples up to 1 kg can be introduced by a pneumatic conveyor which connects a storage cell with the process cell line.

The equipment will be mounted outside the cell on movable floor panels. These panels can be introduced into the cells by use of a caisson which - after removal of the concrete plug - can be flanged to the rear frame of the containment box. Using a double-cover technique, the rear panel of the cell and the door of the caisson are lifted into the caisson and the equipment is pushed or pulled into the cell cubicle.

2.4 Utility lines

Each cell is equipped with a cooling water circuit, compressed air and vacuum lines, all of them controllable by motorized and electromagnetic valves. The electrical plugs (gold plated LEMO-plugs) are α -tight (indium wire and rubber gaskets).

2.5 Ventilation

Usually, the air in the cell cubicle is changed 36 times per hour (Fig. 6); in emergency cases, the ventilation rate can be increased to a 50 fold change.

The air is filtered, but not recycled or cooled; during normal operation of all the illumination lights (4 x 60 W sodium lamps, 2 x 500 W iodine lamps) the air temperature in the cubicle does not exceed 30°C.

2.6 Waste disposal

Solid waste is disposed of by use of a shielded container accessible through the transfer port in the rear plug (Fig. 7). Liquid waste is collected in the basement in shielded glass bottles from which the highly active solutions can be pumped into a shipping container.

3. Equipment

Our first processing is based on the standard procedure developed in the U.S.^{3,4} It consists of the following steps :

- dissolution of the slug in sodium hydroxide and nitrate solution,
- separation of the supernatant by centrifugation,
- dissolution of the residue by aqua regia,
- sorption of the plutonium from 6 M hydrochloric acid on Dowex 1 resin,
- sorption of trivalent actinides and lanthanides from 12 M lithium chloride, and their
- separation by elution with 10 M lithium chloride - 0,1 M hydrochloric acid in presence of methanol.

Pyrex glass will be used. Whenever possible, metal was substituted by plexiglass or polyvinylchloride; ball joint clamps are coated with plastic.

3.1. Dissolver vessel.

The dissolver vessel (Fig. 8) is connected to a reflux condenser, a set of absorption towers and a sucking pump. The subpressure can be regulated and measured with a manometer inside the cell. The water bath is heated by a quartz-covered heating rod; thermal equilibrium is achieved by bubbling air through the water bath.

3.2. Transfer Pipetter.

After complete dissolution of the slug - we hope not to be obliged to dissolve in hydrochloric acid -⁴, the alkaline slurry will be transferred into centrifuge tubes. Using a transfer pipette and a remotely controlled Methrohm-buret, volumes up to 50 ml can be transferred. (Fig. 9)

./..

3.3 Evaporator

After centrifuging and redissolution of the hydroxides, the solution has to be evaporated. The centrifuge tubes are heated in a heating jacket, while hot air is blown on the surface of the solution (Fig. 10). Like the dissolver vessel, the evaporator is connected to a condenser, a set of absorption towers, a liquid scrubber and a pump.

3.4 Column Stand

The heating jacket of the ion exchange column is heated by a commercial solder iron heater. Air pressure is applied to the top of the column. Regulating valve and manometer are inside the cell. The fraction collector can be turned by the manipulator (Fig. 11).

3.5 Solid State Detectors

The elution of the activities from the column is checked by solid state detectors, similar in principle to the ones used in Berkeley.⁵ Commercial ORTEC-detectors are connected to slightly modified FRIESECKE-HÖPFNER monitors and discriminate between α and β radiation. The detectors are protected by mylar foil.

3.6 Micropipetter

There is no direct connection between the cell and the operating area. Therefore, the micropipetter (Fig. 12) has to be operated by manipulators. A microsampling device recently developed by "PUMPETT" (Pumpett 18 Mikro, Ingeniörsfirma Pumpett, Åby, Sweden) is connected to the micropipette and fixed with a plastic ball joint. The centrifuge tubes and the sample bottles can be positioned by a turntable and a small laboratory jack. Sampling from 10 μ l to 1 ml is possible.

3.7. Tests.

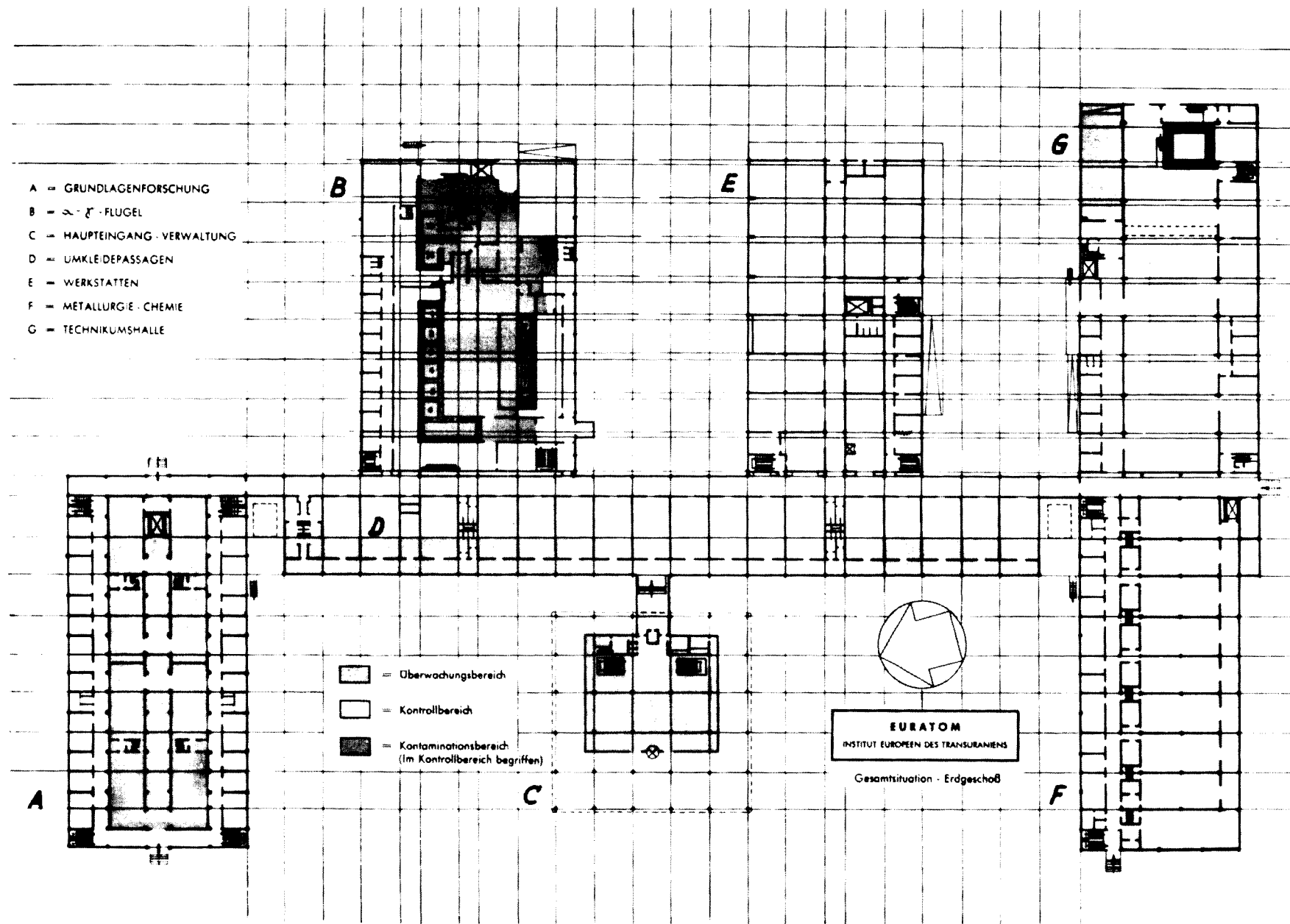
The separation of americium and curium, with both synthetic mixtures of ^{241}Am and ^{244}Cm and with the ^{243}Am - ^{244}Cm fraction isolated by KOOI, served to check the column and detector performance.

The actinides were sorbed from 8 M lithium nitrate on a kieselguhr column loaded with tridecylmethyllumonium nitrate (aliquat 336), then eluted by 4 M lithium nitrate solution. The composition of the fractions was analyzed by α - or mass spectrometry. Our results confirmed the excellent behavior of extraction chromatography systems involving quaternary ammonium salts as shown by HORWITZ et al.⁶ at Argonne and by VAN OUYEN⁷ at Petten/Netherlands. The other material could only be checked in a cold run, since the official approval for the use of the hot cells has been obtained only on November 1st. The actual processing will begin after this Symposium.

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- 7) J. VAN OUYEN, paper presented at the International Conference on Solvent Extraction, Gothenburg, (Sweden), August 27-September 1, 1966.

Fig. 1 - European Transuranium Institute - First Floor Plan



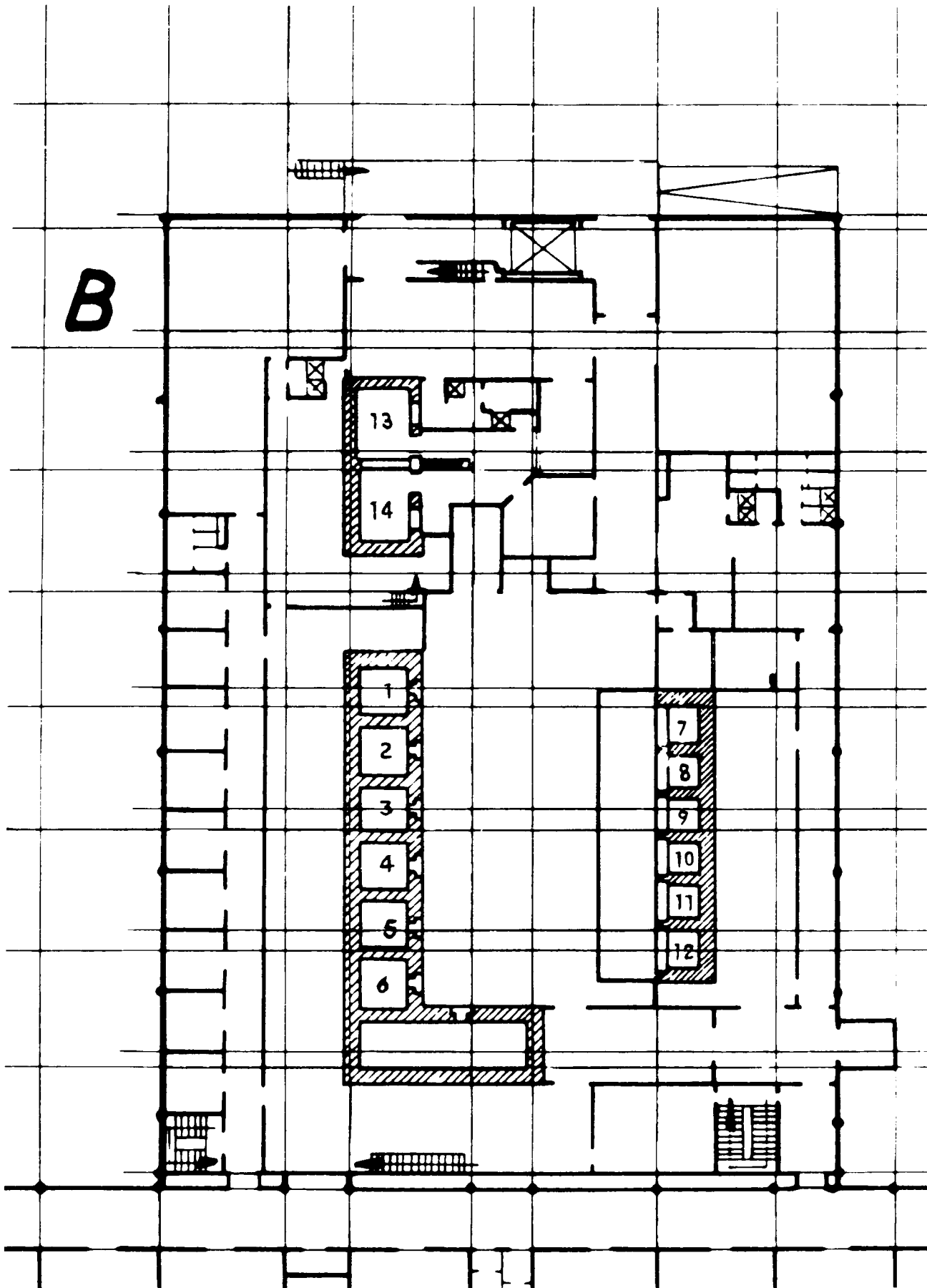
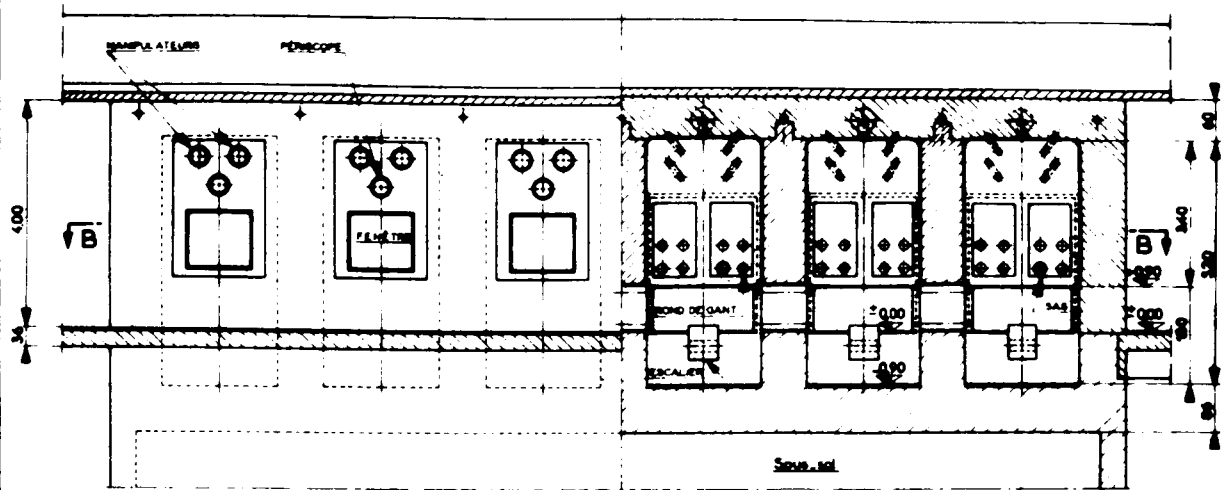


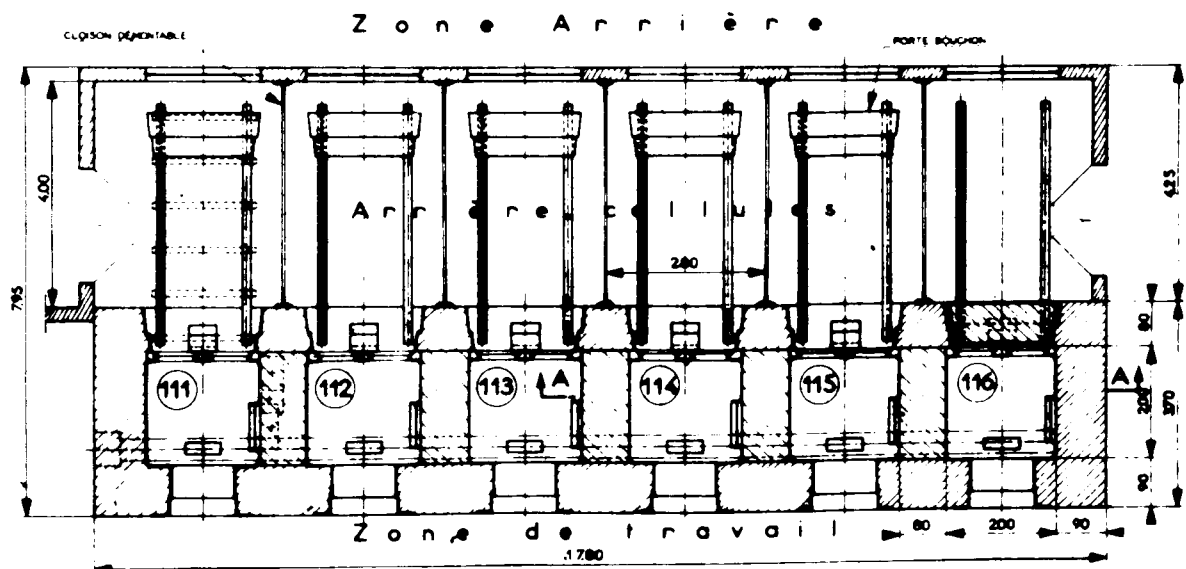
Fig. 2 - Hot Cell Wing

1/2 FACE AVANT

1/2 COUPE A



COUPE B.



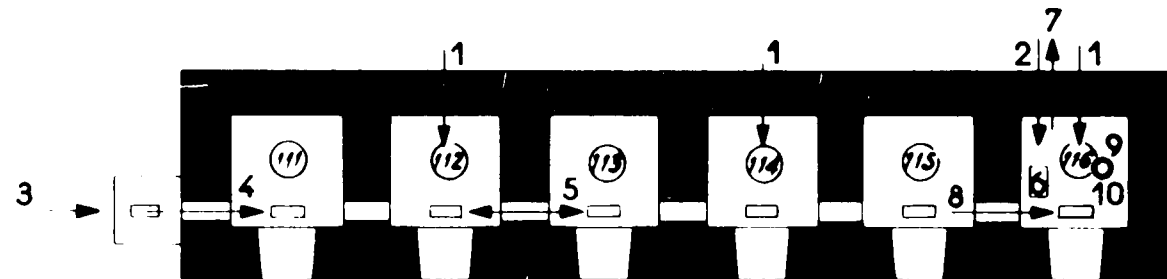
PETITES CELLULES.

ENSEMBLE

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PLANCHE 33

Fig. 3 - Chemistry Cells

Fig. 5 - Transfer of Material



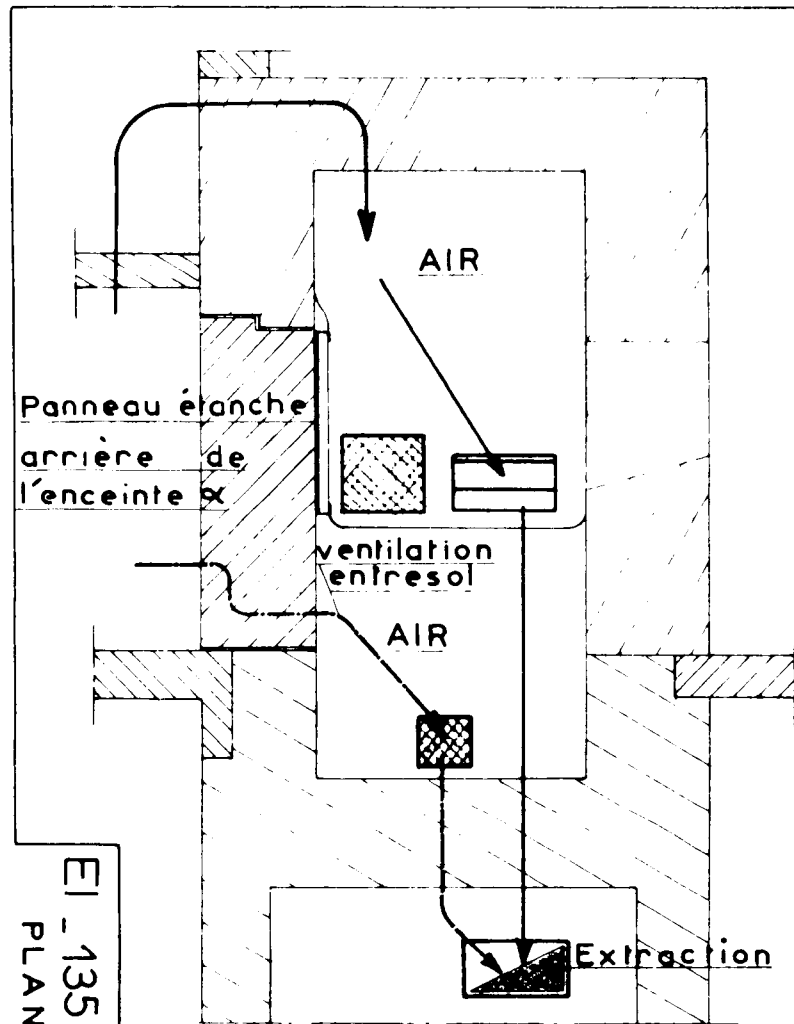
PETITES CELLULES

Transferts

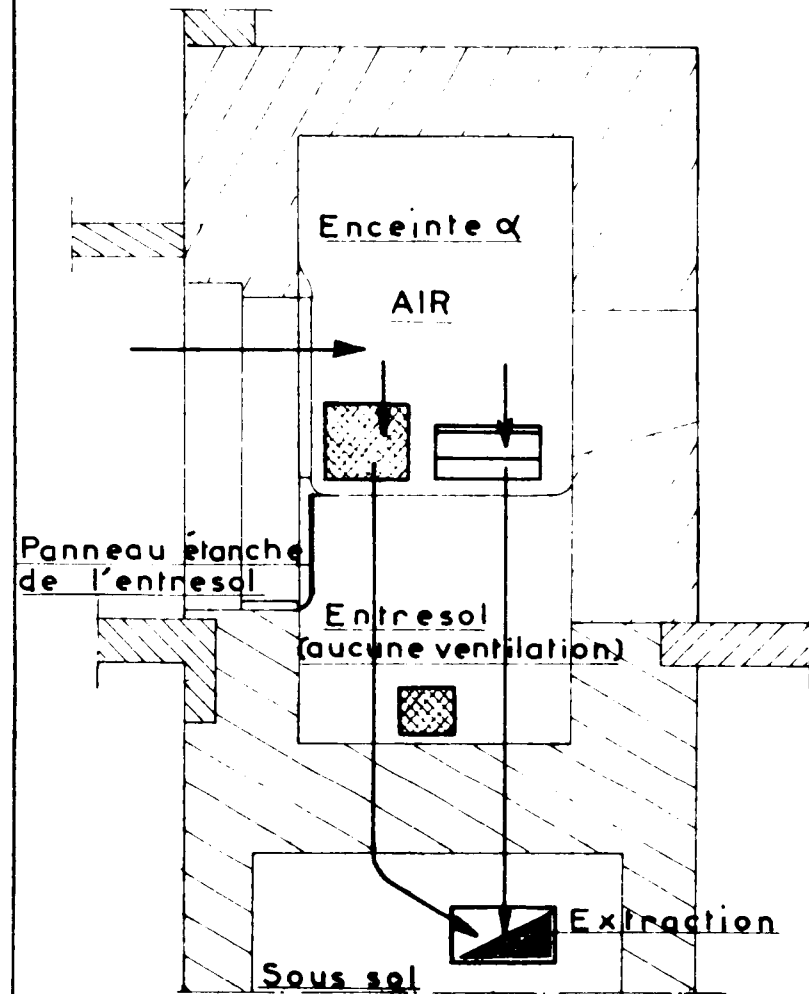
- 1 Introductions des matériaux irradiés par chateaux
- 2 Introduction des matériaux irradiés par ^{de plomb}
- 3 Introduction des matériels froids ^{transfert pneum.}
- 4 Transfert des matériels froids
- 5 Transfert par convoyeur des matériaux irradiés
- 6 Stockage des matériaux actifs
- 7 Transfert vers grandes cellules
- 8 Transfert des déchets
- 9 Sortie α des déchets
- 10 Sortie β des déchets

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PLANCHE 35

Fig. 6 - Cell Ventilation



1^{er} cas : Porte bouchon fermée

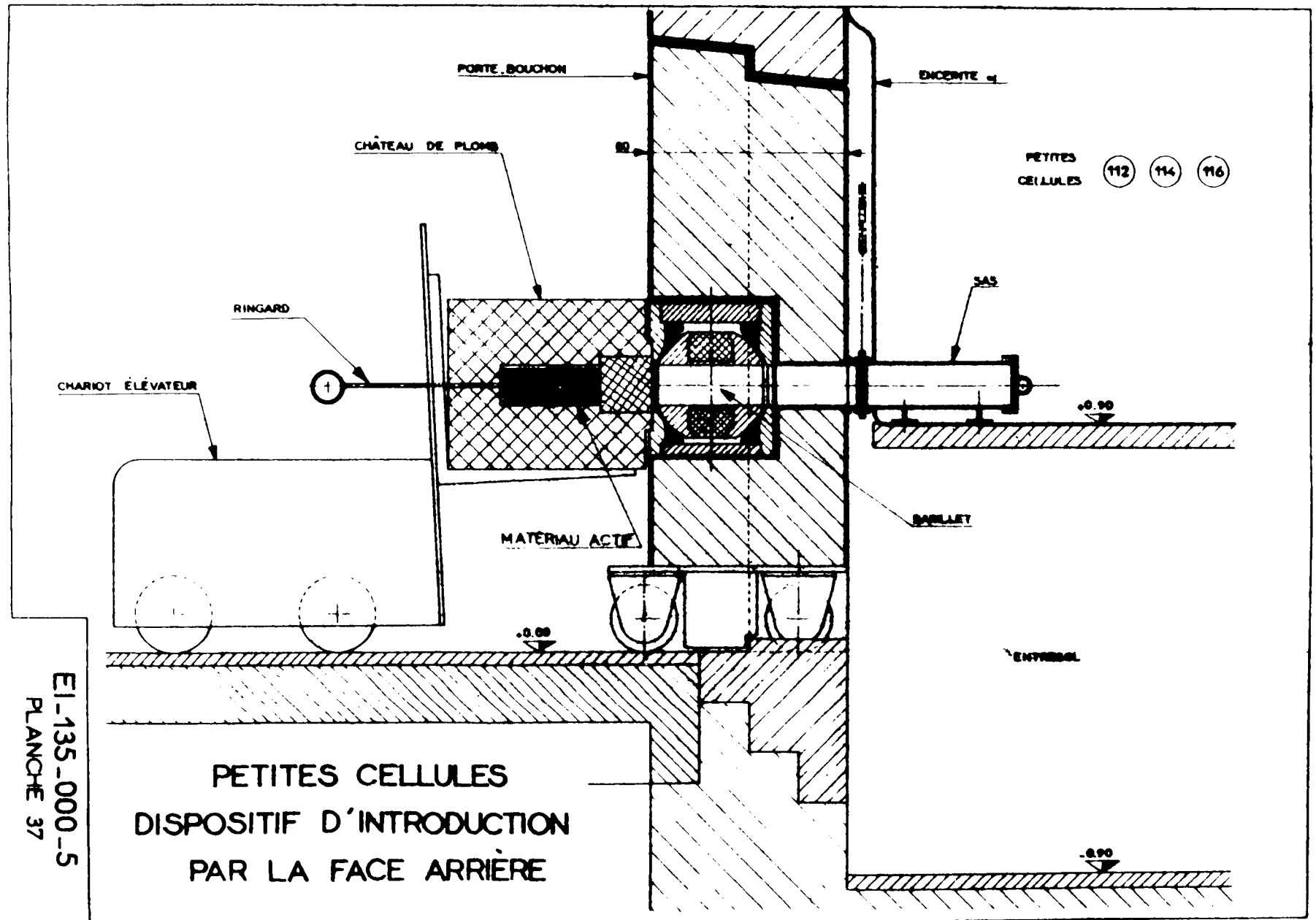


2^{es} cas : Porte bouchon et enceinte α ouverte

VENTILATION EN AIR DES PETITES CELLULES

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PLANCHE 49

Fig. 7 - Solid Waste Disposal



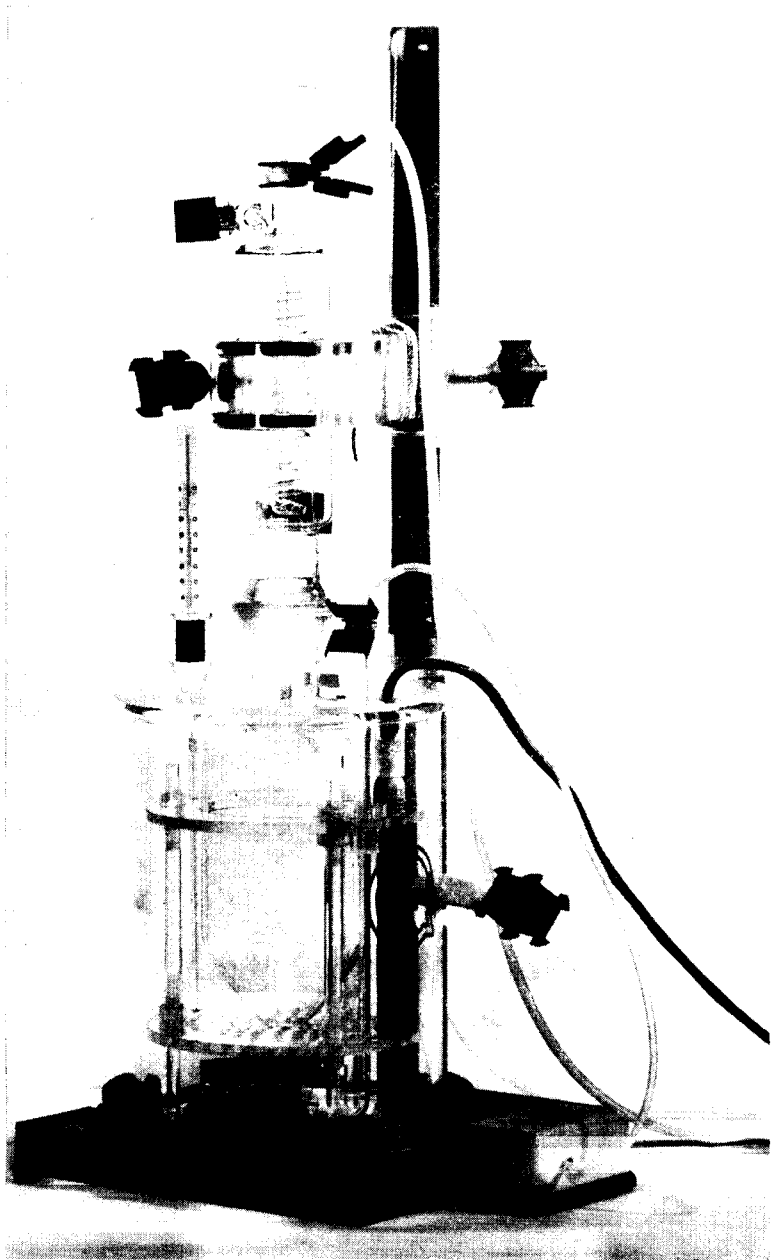


Fig. 8 - Dissolver Vessel

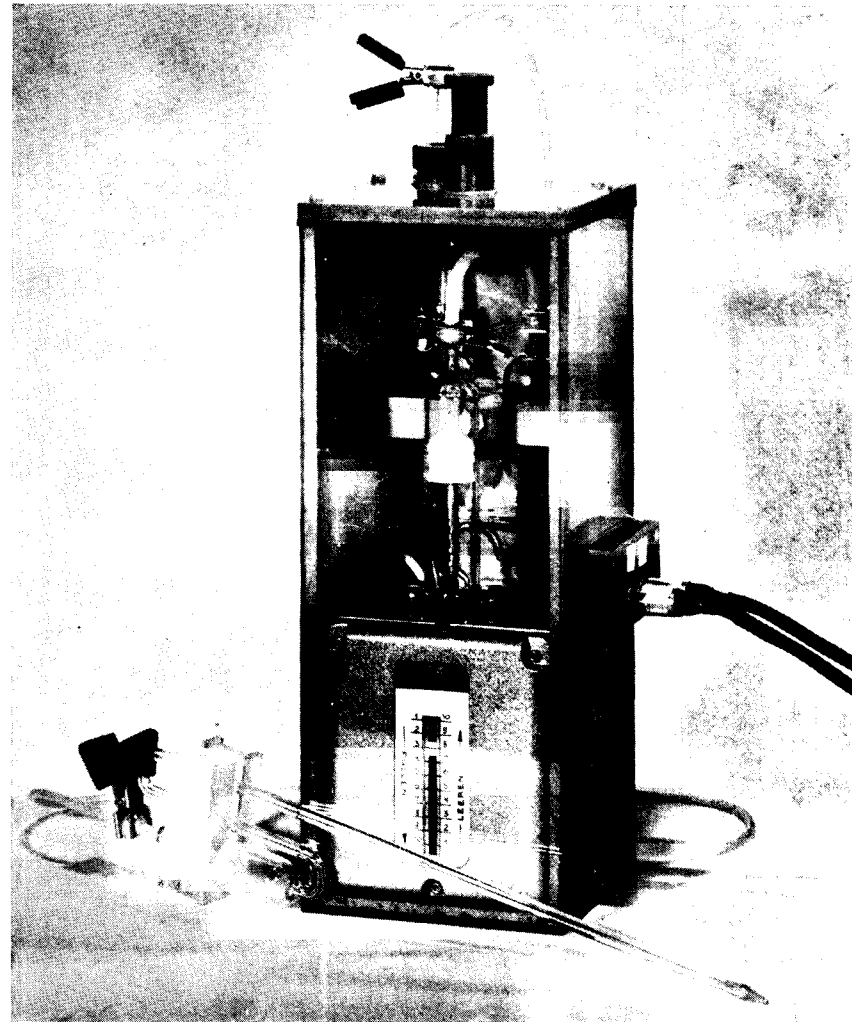


Fig. 9 - Transfer Pipetter

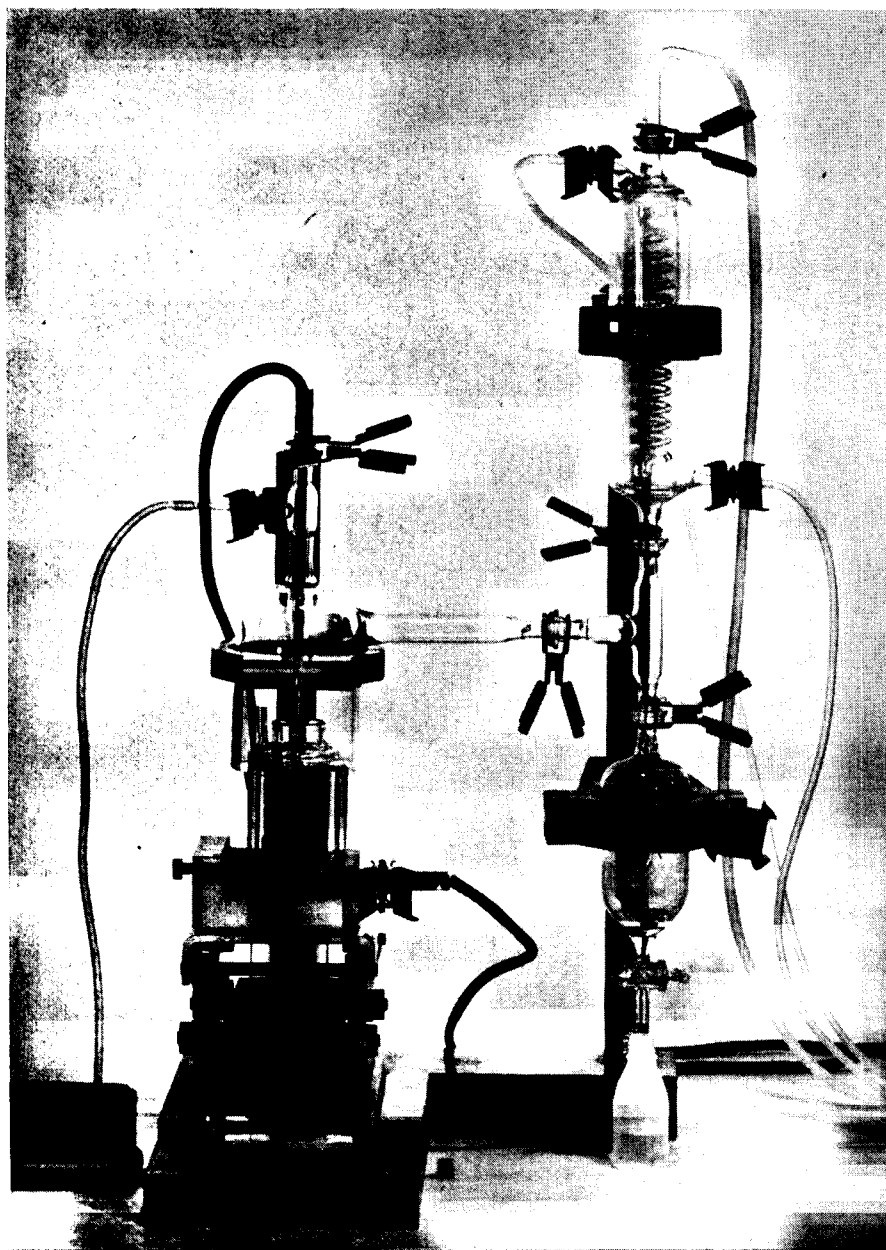


Fig. 10 - Evaporator

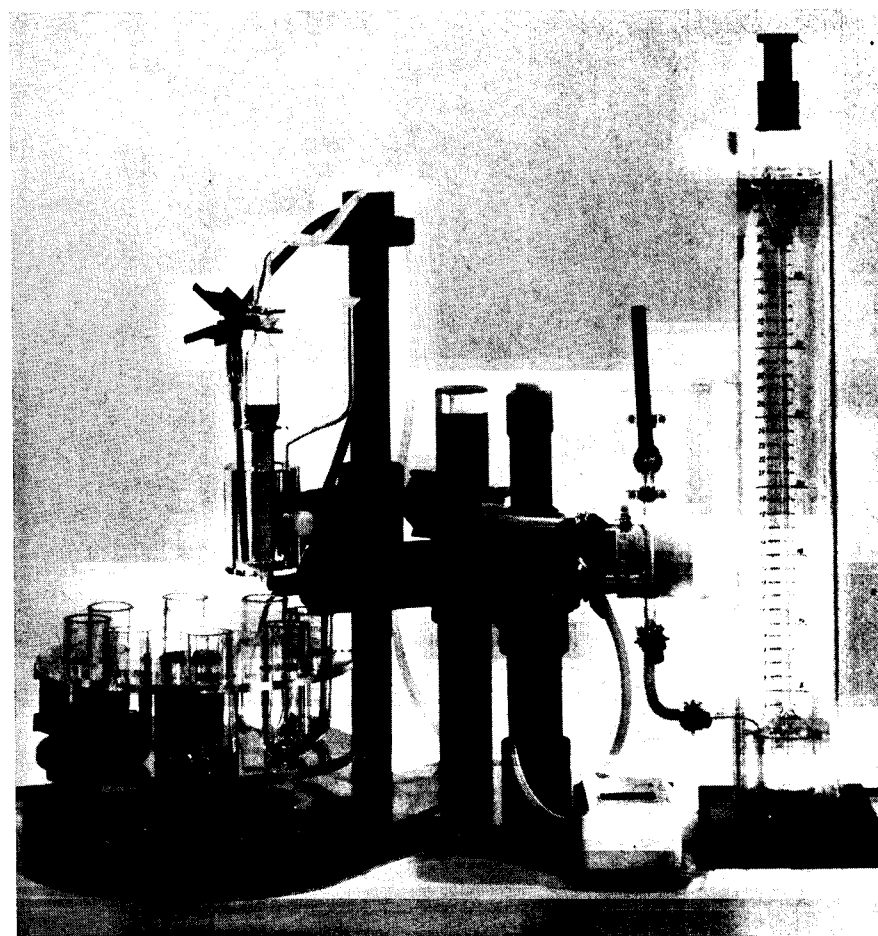


Fig. 11 - Fraction Collector

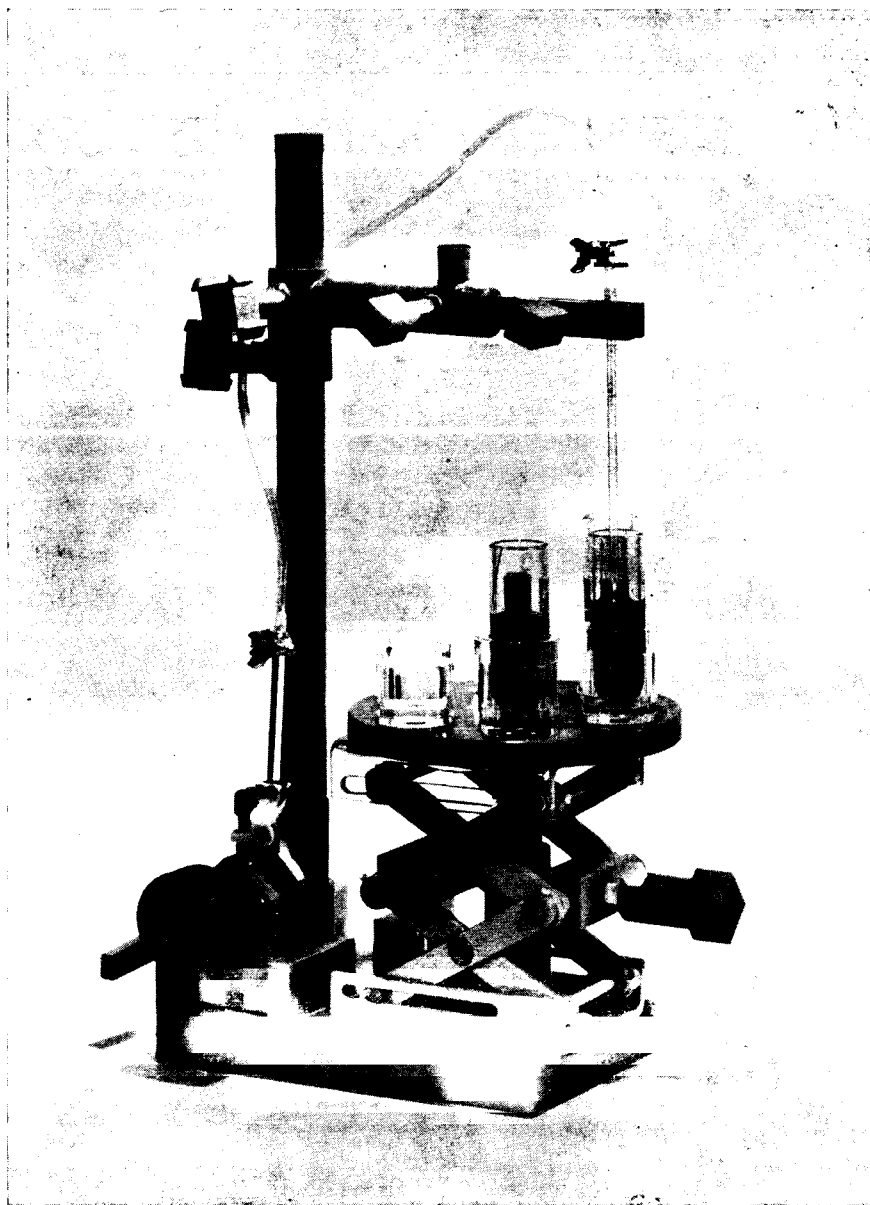


Fig. 12 - Micropipetter

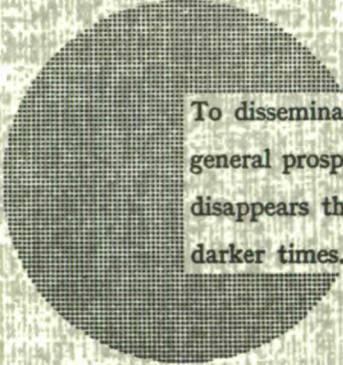
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To disseminate knowledge is to disseminate prosperity — I mean general prosperity and not individual riches — and with prosperity disappears the greater part of the evil which is our heritage from darker times.

Alfred Nobel

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