COMMISSION OF THE EUROPEAN COMMUNITIES

nuclear science and technology

The Community's R & D Programme on radioactive waste management and storage

(Second annual progress report)

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Directorate General for
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F O R E W O R D

This is the second progress report concerning the programme on radioactive waste management and storage (indirect action) of the Commission of the European Communities. It covers the year 1977 and follows the 1976 Annual Report.

The five year programme, ending 31 December 1979, was approved by the Council of Ministers on 26 June 1975.

The Council decision is based on the two following considerations[+]:

- "Nuclear energy is bound in the near future to become one of the main sources of energy alongside traditional sources, and its specific nature requires permanent monitoring of its potential effects and improved measures and research to protect the environment;

- The development of nuclear energy inevitably involves the production of radioactive waste and it is therefore essential to find effective means for ensuring the safety and protection of both man and his environment against the potential hazards involved in the management of such waste".

The programme is carried out by contracts on an expense sharing basis with public or private qualified agencies of the Community; the Commission's financial participation amounts to 19,16 million units of account[++]. A Consultative Committee of national experts nominated by their governments assists the Commission for the management of the programme.

The aim of the programme, as indicated in the annex of the Council decision, is the joint development and perfecting of a system of management of radioactive waste produced by the nuclear industry which, at its various stages, affords man and his environment the best protection possible.

[+] O.J. Nr. L 178/28, 9.7.1975

[++] 19,16 Mio u.a. = approx. 23 MioUS $ on 1.1.77.

The principal items of the programme are the following :

A. Work to solve certain technological problems posed by the processing, storage and disposal of radioactive waste.

Processing :

- Medium activity solid waste : coating with plastic resins;
- High-activity solid waste : decontamination and conditioning of irradiated fuel element cladding;
- High-activity solid waste : immobilization of calcined waste from fission products in a metal matrix;
- Plutonium-contaminated solid waste : incineration process;
- Comparative study of the properties of various materials suitable for the immobilization of high-activity waste.

Storage and disposal :

- Storage of solidified radioactive waste in engineered structure;
- Disposal of radioactive waste in suitable geological formations, including those formations currently being studied;
- Storage of gaseous waste.

Study of an advanced management model :

- Separation and recycling of long-life waste (actinides).

B. Work contributing towards the definition of a general framework (legal, administrative, financial) for the implementation of radioactive waste storage and disposal measures :

  - Review of problems posed by the management of radioactive waste which could not be solved under existing international, legal, administrative and financial provisions and proposals for solutions;
  - Study of principles which should govern the management of radioactive waste.

The professional staff in charge of the management of the programme during 1977 were :

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CEC
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with the assistance of C. Cottone, W. Krischer and W. Falke

The programme is closely coordinated with the activities related to the radioactive waste management conducted by the Common Research Center of the CEC within its pluriannual Research programme (direct action).

S. Orlowski
Head, division
"Nuclear fuel Cycle and
power industry"

C O N T E N T S

1 TREATMENT OF LOW AND MEDIUM LEVEL WASTES

1.1. Introduction

Liquid wastes from virtually all operations of the nuclear fuel cycle against immobilization for safe transportation and storage.

The low and medium liquid level waste arising from LWR largely consists of evaporator sludges, ion exchange resins and filter aids. Similar wastes are produced in certain operation of reprocessing plants. The presently applied immobilization techniques, using concrete or bitumen as metric, have not been entirely satisfactory. In certain cases solidification of the concrete was not assured or the waste volume was considerably increased. Bitumen embedding, among other shortcomings, has off-gas problems, uses an inflammable matrix and produces solidified wastes with low mechanic strength.

The immobilization in thermal setting resins, e.g. polythene, polyester or epoxy should at least for a wide range of low and medium level liquid wastes render waste products with improved characteristics for transport and disposal.

1.2. Mobile Pilot Plants for Resin Embedding of Reactor Wastes (Steag Kernenergie Gmbh)

1.2.1. Objective and Scope

Steag Kernenergie Gmbh, the contractant, was and still is the only firm to commercially condition radioactive bead ion exchange resins by embedding this type of reactor waste in polystyrene resins. The objective of the R & D Contract in this programme was to study the possibilities and develop a technology permitting the embedding of other reactor wastes (e.g. evaporator sludges, filter candles, powder resins) in a suitable polymer matrix.
The programme was intended to proceed in 5 stages :

a) preliminary investigations (product criteria, characteristics of the wastes);
b) study and choice of candidate resins and conditioning methods;
c) laboratory studies and technical experiments with inactive and active wastes;
d) operation of experimental pilot plants;
e) data processing and analysis of the results.

1.2.2. Preliminary Investigation

The following investigations were carried out :

a) The various types of reactor wastes were reviewed in order to determine the range of wastes for which the resin fixation technique was advantageous;
b) A set of preliminary requirements and conditions of acceptance for storage and disposal of the product was drawn up;
c) Safety and operational characteristics of the immobilization substances were studied and a first list of criteria was compiled;
d) The various types of resins were examined against these criteria and exploratory tests with inactive wastes were undertaken.

1.2.3. Results

a) Reactor wastes

Of the wastes produced by light water reactors : solid incombustible, solid combustible and wet or liquid wastes, only the latter category should be considered as most of the solid incombustible wastes (absorber and control rods, in core measuring devices etc.) are normally too active for organic polymers and the solid combustibles : e.g. contaminated clothing and cleaning material are generally of such low activity, that they can be conditioned and stored without fixation. The low active liquid or wet wastes can generally be conditioned, but almost all require drying prior to incorporation into resins. The amounts of these wastes produced by the modern large LWRs are, according to a STEAG survey, somewhat lower than assumed in some of the recent literature.
The survey shows that a 1200 MWe PWR produces only about 50 m^3/a of low active wet waste in form of filter aids and evaporator sludges and 1-3 m^3/a of medium active spent ion exchange resin.

b) Quality and Safety Criteria of the Product

The most important characteristics for LLW and MLW immobilization products were found to be :
- high chemical stability of the immobilization substance against water and various salt solutions;
- high leach resistance of the waste product;
- stability against thermal effects;
- chemically and biologically safe decomposition products;
- no liquid or gaseous release;

- low flammability;
- no violent chemical reaction;
- high resistance against bacterial decomposition, fermentation and other forms of decay.

The second group of characteristics, listing second priority properties was also drawn up. These included such physical properties as softening point, density, mechanical strength, thermal conductivity and homogeneity of the product.

c) Safety and ease of handling of the bonding resins

In addition to the waste product criteria the polymers or other immobilization substances should be safe and convenient to handle in a reactor waste treatment plant.
The main requirements in these aspects appear to be :

- low viscosity;
- low volatility;
- high flame point;
- low adhesion tendency;
- low toxicity;
- low specific reaction heat;
- good control for the polymerisation;
- good stability during storage.

d) Choice of resins

First series of tests with polystyrene, polyester and epoxy resins have been carried out but numerical results are not yet available. A more qualitative comparison of the three products is presented in table 1.

Property of immobilizing resin	Polystyrene	Polyester	Epoxy
Leaching resistance (hardened)	High	high	high
Fire hazard	high	high	low
Reaction heat	high	medium	low
Volatility	high	medium	low
Toxicity	high	high	low
Viscosity	low	high	high
Capacity to absorb waste moisture	high	low	low

Other products - polyuremethanes and resin - cement systems were also considered. First tests with polyuremethane confirmed that the leach resistance of this type of polymer is comparatively poor.
The cement-polymer systems however promise to have all the advantages of the cement fixation (low fire hazard, good moisture absorption) without the disadvantages (mechanical strength, leach resistance).

1.2.4. Conclusions

The polystyrene resin embedding, originally favoured by Steag, does not, in the view of the contractor have a sufficient development potential to warrant further effort on this contract. The main reasons for this conclusion are :

- Handling problems with the raw resins (fire and explosion hazard of styrene, toxicity);
- Processing difficulties : nearly all wet reactor wastes require preparatory treatment, generally drying, in order to obtain a satisfactory product;
- Economics
  The progress achieved in improving cement conditioning methods and their products seriously threatens the economic viability of the competing resin based conditioning systems at least in the FRG, where the disposal space in the ASSE Salt mine is not the major cost factor.
- Radiation shielding
  Due to the many process operations, frequent maintenance and often low production rates of the resin conditioning plant, occupational radiation exposure is relatively high.

The Commission and Steag will, in the beginning of 1978, negotiate the discontinuation of the present programme.

1.3. Stationary Resin Embedding Pilot Plant (SENA)

1.3.1. Objectives

The aim of the second R & D contract on low and medium level waste conditioning is to build and operate an active pilot plant for resin fixation of the reactor wastes on the site of the SENA PWR at Chooz and to test and evaluate the characteristics of the conditioned waste product in respect to storage and disposal.

1.3.2. The Development Programme

The R & D programme implemented under the contract consists of the following main actions :

- definition of the conditioning process;
- design and construction of the pilot plant;
- commissioning and operation of the pilot plant;
- testing and evaluation of the products.

1.3.2.1. The Conditioning Process

The process of fixing wet reactor waste in thermo-setting resins as patented by the SENA will primarily be applied to the evaporator sludges. These wet residues will first undergo chemical pre-treatment, in order to fix the principal radioisotopes Cs and Co to insoluble compounds. There upon they are dried in a continuous rotating drum dryer and added to the polymer resin in the final packing drum. The dryer off gas is sent through a scrubber (DF 10^3 to 10^4) and a condenser before entering the reactor off gas system. The mixture is then stirred and the accelerator and catalyst are added to start the setting.
The waste-resin mixture then hardens within hours.

1.3.2.2. The Reactor Wastes

The amount of wastes that will be treated in the pilot plant are :

- ca. 30 m^3/a of primary wastes (50 Ci/m^3);
- ca. 15 m^3/a of miscellaneous wastes (1 Ci/m^3).

The primary wastes essentially consist of sodium borate (200 g/l) and boric acid (130 g/l) and contain the active isotopes Cr_{51}, Co_{60}, J_{131}, Cs_{134}, Cs_{137}.
The miscellaneous wastes will contain 100-200 g/l of various solid residues of detergents, decontamination products, sodium borate etc.

1.3.2.3. The Pilot Plant

A pilot plant applying this CEA process will be built commissioned and operated with active waste on the site of the Chooz PWR. The plant will condition the wastes from the reactors effluent treatment facilities and produce canned waste blocks acceptable for transport and storage.
The construction of the pilot plant is scheduled to be completed in March 1979.

1.3.2.4. Commissioning

At the end of the construction period a series of tests with inactive simulated wastes will provide opportunity for systematic trials at various operating conditions and waste compositions.

1.3.2.5. Operation

The last eight months of the programme will be devoted to active operation under representative conditions.
All operating parameters, inspection and maintenance results as well as occupational exposure to radiation and, if any, toxic substances will be recorded and evaluated.

1.3.3. Product Characteristics

A special study will establish the requirements for transport and storage of the waste product.
Production and, to a certain extent, experimental samples from the pilot plant will be tested and assessed against these characteristics.

The product evaluation study will also compare the relevant properties of the SENA product with those achieved with cement and bitumen on similar wastes.
Statistic analysis of the quality control of the product will provide information on the effects of variations in operating conditions and on the consistency of the production.

1.3.4. Present State of Advancement

Work on preparing the site has begun in summer 1977 and, at the end of 1977, building work was reported to be progressing to schedule.

The principal target dates for the work in 1978 are :

Completion of project definition and of technological tests : 31 May 1978;

Completion of building and civil engineering : 30 October 1978;

Commissioning of the pilot plant : 30 November 1978;

Operation of the plant : 30 March 1979.

2 HIGH-ACTIVITY SOLID WASTE : DECONTAMINATION AND CONDITIONING OF IRRADIATED FUEL ELEMENT CLADDINGS

2.1. Introduction

Cladding wastes, whose characteristics are given in table 2.1. consist of cladding and structural components of the fuel assemblies that remain after chop-leach operations at the reprocessing plant.

Table 2.1 : Unconditioned cladding wastes from LWR spent fuel

Waste composition (d = 1)	Arising from 1400 MT/y reprocessing	Radioactivity (1)	Thermal power (1)
- Zircaloy 75 % - Stainless steel and Inconel alloy 25 w/o	560 m^3/y	150 kCi/m^3	850 W/m^3

(1) Burn up of 33 GWD/MT; cooling time of 1 year; contamination by 0.5 % of total spent fuel.

Conditioning processes, combining, if possible, an appropriate immobilization of radioactivity for long-term disposal with a substantial volume reduction, are needed for this waste.

2.2. Press Compaction and Encapsulation by Low-Melting Alloy (CEN/SCK Mol)

Assembly extremities are assumed to be regarded as a non-waste. The rest of cladding waste will be mechanically compacted into blocks and placed in a special basket. The remaining voids will be filled up with a corrosion resistant lead alloy by casting at about 723 °K. The research programme for the development of this conditioning method has been already described (1).

2.2.1. Assessment of Filling Alloys

Lead-tin and lead-antinomy alloys were selected as candidate filling materials. The corrosion resistance of those alloys may be improved by small additions of tellurium or calcium.

2.2.1.1. Corrosion of Filling Alloys in Disposal Environment

Potential alloys have been submitted to tests in which the wet atmosphere of an underground storage in a clay

formation has been simulated. The results in water environment (table 2.2) show the smallest corrosion rates in the case of ground water whose impurities form a protective layer or act as corrosion inhibitors.

Table 2.2 : Weight changes (mg/cm^2) of lead alloys after exposure to different environments at 322 K.

Alloy	Ground water (+)		Demineralized water			Humid clay atmosphere
			Saturated with air		Absence of air	
	15 days	3 months	15 days	3 months	2.5 months	1 month
Pb	- 0.037 - 0.035	- 0.018 - 0.059	- 18.18 - 17.79	- 33.99 - 35.51	+ 0.626 - 6.67	+ 0.14
Pb-Zr-Mg	+ 0.186 + 0.185	+ 0.186 + 0.148	- 13.08 - 18.59	- 18.48 - 24.58	- 5.87 + 2.27	+ 0.29
Pb-Te	0 - 0.072	0 - 0.062	- 35.75 - 33.28	- 52.32 - 49.80	- 9.74 -11.73	+ 0.14
Pb-Sb	0 - 0.019	0 + 0.069	- 16.76 - 16.38	- 26.15 - 28.19	+ 1.93 + 1.16	+ 0.14
Pb-Ag	0 + 0.019	+ 0.29 + 0.24	- 6.76 - 2.64	- 9.11 - 13.18	-39.00 -39.02	+ 6.83

2.2.1.2. Interaction of the Filling Alloy with Cladding Waste and with Canister Materials

The compatibility of a Pb-1.5 w/o Sb alloy with zircaloy 4 and AISI 304 (potential canister materials) is being evaluated after casting under vacuum at 723 K and long-term heat treatments up to 373 K. Attack of AISI 304 at the interface is caused by alloy impurities such as Cu, Ag, Zn and Cd. No reaction was observed in the case of Zircaloy-4.

2.2.1.3. Influence of Filling Alloy on the Mechanical Properties of the Canister Material

A lead surface coating has been applied on three candidate canister materials (carbon steel, AISI 316 and AISI 430) for the experimental study at temperatures up to the melting point of lead.

2.2.2. Development of the Conditioning Process

2.2.2.1. Compaction

Compaction tests have been performed by means of an extrusion press on unirradiated stainless steel and zircaloy-4 cladding tubes. The results of these tests are graphically represented in fig. 2.2.1. The compact diameter and the filling height (Ho) only have a minor influence on the obtained volume reduction.

Fig. 2.2.2 shows a typical example of compacted zircaloy tubes.

2.2.2.2. Embedding in Low Melting Alloys

Embedding tests of compacted zircaloy chops have been performed with Pb-1.5 % Sb as filling alloy by air and by vacuum casting at 723 °K. Vacuum casting (fig. 2.2.3) results in a much better filling (1.5 - 3.5 % rest porosity) in comparison to air casting (15 - 18 % rest porosity). Generally a good contact between zircaloy and lead alloy has been observed and no open porosities or crevice propagation have been found.

2.2.3. Conceptual Development

A preliminary concept has been based on a compact unit with capacities from 1500 to 3000 tons/a and producing compacted waste briquettes of 34 cm diameter and 6 cm height. About 3600 loadings have to be compacted annually by a unit of this size in a 300 MTU per year reprocessing plant which would require a smaller press.

Press constructors have been contacted to evaluate the realization of a press unit in a hot cell at reasonable costs.

A conceptual design of a waste canister, which can be used as mould for casting of filling alloy, has been completed (200 cm length, 35 cm internal diameter, 3 mm wall thickness).

2.3. Conditioning by Rolling and Embedding in Concrete (KFK - Karlsruhe)

The method consists in incorporating in concrete the hulls whose volume is reduced by rolling press and the structural materials. The process will be developed and optimized by inactive tests at a pilot plant. The research includes the conceptual study of a reference process, as well as a comparative study of the various processes for treating cladding wastes.

2.3.1. Process Development and Pilot Plant Conception

The rolling characteristics of inactive hulls were studied at a laboratory press. Zircaloy-4 hulls (10.75 x 0.72 x 50 mm) may be compacted till about 45 % of their theoretical density by single piece feeding with a deforming force of not more than 16 kN. After rolling three times about 30 % of hull pieces were longitudinally broken. 0.03 % fines were produced. The design of a half scale compacting unity (fig. 2.3.1) for cold tests has been started. The cladding wastes are discharged from a 400 liter container (A) into the feeding hopper (B). By means of a vibrating runner (C) the structural pieces, which are directly conveyed to the drum (G) containing the fluid concrete, are separated from the small pieces and hulls. The latter will, one by one, reach the compacting rolls (E) through a dosing gear (H). The compacted pieces will then fall into the drum (G).

2.3.2. Examination of Conditioned Wastes for Disposal

2.3.2.1. Hydrogen and Water Release

Physical and chemical reactions were examined on various concrete and zircaloy (fines)- concrete mixtures, assuming that the waste temperature does not exceed 100 °C. Some tests were extended up to 400 °C to consider accident conditions.

Hydrogen is released by :

- Thermal effect

  Different concrete mixtures were tested at temperature up to 400 °C and the results condensed in a graph are given in fig. 2.3.2. Concrete with additions at 200 °C already produced a H_2 release rate, which may lead to an inflammable and explosive gas mixture.

- Water zirconium reaction

  Zirconium shows a high reactivity with the water present in concrete. Measurements were carried out on H_2 release from a zirconium fines-concrete mixture as a function of temperature, zirconium content and duration of the experiment (fig. 2.3.3). Concrete additions seem to favour the reaction.

- Water radiolysis

  The water present in concrete undergoes the following radiolytical reaction :

$$3\ H_2O \rightleftharpoons H^\circ + OH^\circ + H_2 + H_2O_2$$

  Concrete samples (10-15 g) sealed in glass (Argon atmosphere) were irradiated to gamma dose of 20,

50, 100 and 200 Mrad.
The first results indicate H_2 releases of 2.2 - 6.5 µl/g concrete per Mrad corresponding to 40 - 70 % of the produced hydrogen.

- Water

Water may be eliminated by thermal effect. Laboratory tests gave weight losses up to 26 % after 1 day at 100 °C. The results are given in table 2.3.

Table 2.3 : Water release from concrete samples at 100 °C.

CONCRETE	EPZ-350	PZ-350F	PZ-450F	PZ-450F + 1 % Tricosal	PZ-450F + 1 % Addiment	Blitz-dämmer
Weight loss (w/o)						
After 3 h	7.2	2.2	5.3	7.6	2	8
9 h	19.3	11	16	15.3	10	18
24 h	26	21	22	19	17	22

2.3.2.2. Tritium Release

Assuming a fuel burn up of 33 GWD/te, a cooling time of 3 years and a 60 % tritium retention in zircaloy, hull wastes should have a tritium activity of 1 Ci/kg Zr. Isotopic exchange reactions in the system zircaloy-tritium-concrete-water were investigated in following media :

- Hydrogen produced by water radiolysis during irradiation at 100 °C to gamma doses of 20-100 Mrad and
- Water at 21 °C and 100 °C
- Aqueous solutions with pH 1-10 at 100 °C

Samples were prepared with tritiated zircaloy (0.1 - 0.15 mCi/g Zr).

As yet there is no evidence of isotopic exchange phenomena. Tritium release seems caused by thermal effect. Thermal tritium release values in the temperature range 100-400 °C are given in table 2.4.

Table 2.4 : Tritium release from concrete overcoated tritiated zircaloy hulls at various temperatures and experimentation time.

Sample Nr	Temperature (°C)	Experimentation time (h)	Relative release rate (Ci/Ci · day)
2; 3; 4	100	24	$4.1 - 6.7 \times 10^{-5}$
6	300	24	3.6×10^{-3}
	300	150	9.8×10^{-4}
8	400	24	1.1×10^{-2}
	400	150	2.1×10^{-3}

2.3.2.3. Mechanical Stability of Conditioned Wastes

The compression strength has been determined on specimens of hulls (10.8 x 0.8 x 50 mm) incorporated in concrete (PZ - 350 + 1 % Tricosal) :

Uncompacted Zircaloy	4215 N/cm^2
Compacted Zircaloy	2880 N/cm^2
Concrete specimen (as reference)	3490 N/cm^2

2.3.2.4. Thermal Conductivity of Conditioned Wastes

The thermal conductivity of the heterogeneous concrete-waste system was theoretically estimated for different compaction grades and orientations of the hulls. At a compaction of hulls to 45 % of their theoretical density the thermal conductivity values were in the range 0.057 - 0.064 $W. cm^{-1}. K^{-1}$.

These estimates will be verified by measurements.

2.3.3. Preliminary Study of the Reference Process

Assuming compaction of the hulls to 45 % of Zircaloy density (objective) a 200 litres waste drum will contain 560 kg of compacted hulls and structure materials in a concrete matrix of 210 kg. This corresponds to a volume reduction factor of 2.8 and to a production of 1060 waste drums per year for the conditioning of cladding wastes from a 1400 te/y reprocessing plant.

The alternative of 400 litres drums will be examined. An interim storage for periods of 25 years will be needed for reason of waste management as well as for removal and lowering of heat generation. Considering the high tritium content of hulls, further investigation on its release are needed.

A preliminary assessment shows that the volume reduction of cladding waste will reduce the total costs by 40 % - 60 %.

2.4. Decontamination and Conditioning by Fusion (CEA Marcoule)

Two different processes have been considered for this study :

- Volume reduction by utectic fusion of the waste and decontamination by the introduction of molten glass into the metal bath. Separation of the slag from the alloy and conditioning of both products for the long-term storage;
- Embedding of waste in glass, used elsewhere for fission products solidification;

Most of the work envisaged involves the first process. The original research programme (1) was modified by excluding the conceptual studies of the hot cell and of a reference process for comparative purpose (§2.3.2.4 and chap. 2.3.3 of reference 1). The research work runs from the 1st August 1977 to 31st July 1979.

2.4.1. Laboratory Study

2.4.1.1. Utectic Mixture

Tests have been performed on lowering the fusion points of zircaloy and stainless steel. Different mixtures, each of 200 grams, were maintained for four hours at the selected temperature in argon atmosphere.

Two utectics were considered for zircaloy alloys :

(a) 78.5 w/o Zr - 21.5 w/o Cu at 995 °C;

(b) 83 w/o Zr - 17 w/o Ni at 960 °C.

The fusion tests at 1200 °C on these utectics gave well melted ingots, whose metallographic examinations are under way. Carbon and Silicon were considered as additional material to stainless steel. The results, which are summarized in table 2.5, indicate 1300 °C as the lowest temperature to obtain a complete ingot.

2.4.1.2. Corrosion of Refractory Materials

The ceramics selected for the corrosion study and their composition in weight per cent (values in brackets) are :

(+) ALCOR-B : AL_2O_3 (99.3)

(+) ISOSTAL-G : AL_2O_3 (97 %)

(+) ALUMINE (ISOSTATIC)

(+) ZAC-1681 : AL_2O_3 (48); ZrO_2 (35); SiO_2 (0.5)

(*) ANKER-D1 : MgO (96); CaO (2.5); SiO_2 (1); Fe_2O_3 (0.5)

(*) ELECTREX-F55 : MgO (55); Cr_2O_3 (20); Al_2O_3 (8); Fe_2O_3 (12)

(*) MAGNORITE MN 197 : MgO (97); SiO_2 (1.5); CaO (1.3)

(*) CR_2O_3 - 1215 : Cr_2O_3 (96); TiO_2 (3.6); Fe_2O_3 (0.3)

(*) ZFE : ZrO_2 + HfO_2 (97); CaO (3)

(*) SUPERSTAR : C (25-30); CSi (20-25)

(*) GRAPHITE-ATJ

( ) MAGMALOX : Al_2O_3 (72); SiO_2 (20); ZrO_2 (5); Na_2O (0.8)

(+) C-104 : MgO (58); Cr_2O_3 (28); Fe_2O_3 (7); Al_2O_3 (5); SiO_2 (1.3); CaO (0.7)

(+) CR-100 : Cr_2O_3 (94); TiO_2 (4.2); Al_2O_3 (0.4); MgO (0.4); SiO_2 (0.3)

(+) CSi

(+) CaF_2

2.4.2. Technological Study

2.4.2.1. Prototype Fusion Rig

A simplified prototype rig for preliminary coating experiment at half-industrial scale with simulated claddings, was assembled (Fig. 2.4.1). It consists of a stainless steel box equipped to operate in argon atmosphere and of a tilting electric induction furnace (fig. 2.4.2) with a crucible of 50 kg capacity.

2.4.2.2. Compaction Tests by Fusion

Melting tests in argon atmosphere were carried out on following simulated wastes :

- Zircaloy 4 tubes : length = 20-30 mm
  diameter = 8.25 mm
  wall thickness = 0.5 mm

- Stainless steel AISI 304 L : length = 20-30 mm
  diameter = 5 mm
  wall thickness = 0.5 mm

After fusion of a small part of the material (3 kg), the furnace is fed at a rate depending on the digestion capability of the bath. The main data related to these tests are given in table 2.6.

(*) : For stainless steel and zircaloy alloys casting
(+) : Only for zircaloy alloys casting

Table 2.5 : Preparation tests of stainless steel (18/10) ingots with different additional materials at varying under argon atmosphere

Temperature °C	CARBON					SILICON			
	2 %	3 %	4 %	5 %	6 %	15 %	17.5 %	20 %	25 %
1200						No tests			
1250							n.t.		n.t.
1300	 (30-50% of carbon)	 (~ 30% of C)	 (< 10% of C)	 (20-30% of C)	 (~ 15% of C)				

Total material for each sample = 200 g; dark surface represents the percentage of material forming the ingot; in brackets the amount of additions not incorporated in the ingot.

Table 2.6 : Fusion tests of Zircaloy and stainless steel

Metal	STAINLESS STEEL AISI 304 L					Zircaloy-4
Additional material	-	2 % C	4 % C	6 % C	15 % Si	21.5 % Cu
Bath temperature (°C)	1480	1380	1380	1540	1240	1125
Feeding rate (kg/h)	46	43	35	25	14	50
Ingot characteristics						
- weight	35	34.3	34.7	31	29	22.5
- added material (w/o)	-	1.8	3.75	5.35	+	19.5
- fusion point (liquidus) (°C)	1425	1246	1223	1178	1147	1238
- fusion point (solidus) (°C)	1404	1212	1194	1143	1113	1188

+ Analytical value not yet available.

2.5. Characterization of the Radioactivity in Different Cladding Wastes (UKAEA Harwell)

Analytic examinations of typical irradiated fuel element claddings will be carried out as described in (1). Examinations of two batches of samples were completed (table 2.7).

Table 2.7 : Details on examined batches of samples

Origin of hulls	Batch Nr	Material/sample	Burn up GWD/t	Date of discharge	Fuel
Reprocessing of a PWR (KWO) fuel element at WAK plant	1	Zircaloy	25.5	12.08.71	UO_2 (U-235=2.8%)
Laboratory scale reprocessing of FBR (Dounreay) experimental fuel	4	Stainless steel			$(U,Pu)O_2$; Pu=15%
		SCOF 2	54	06.02.74	U 235 = 93 %
		SCOF 3 (60 V)	43	26.02.74	U 235 = 60 %
		SCOF 3 (93 B)	51	26.02.74	U 235 = 93 %

Hulls were sectioned to provide rings of material which were subjected, directly or after complete dissolution, to the experimental methods.
A detailed description of these methods is given in the literature (2; 3).

2.5.1. Total Content of Actinides

The Pu-239 contents of the hulls were determined (table 2.8) by three different methods :

- Neutron activation

  After irradiation of the samples in a thermal neutron flux for about 1 min and cooling for 30 secs, the delayed neutrons emitted by fission are counted. The method does not differentiate between different fissile nuclides.

- Direct alpha spectrometry

  The content of the main alpha emitter will be evaluated by means of alpha-spectra taken from hull sections themselves.

- Alpha spectrometry of hull solutions

  Actinides were determined by alpha-spectrometry on thin sources prepared from hulls solutions. The Pu-238 and Am-241 activities were measured after separation of Pu (organic phase) from Am (aqueous phase) by 10 % Tri-n-octylamine in xilene.

Table 2.8 : Pu-239 content of hulls (µg 239 Pu/g hulls)

	METHOD		
	Neutron activation	Direct alpha-spectrometry(*)	Alpha-spectrometry on sample solution (1)
Batch Nr 1 (KWO)			
Zircaloy (4 samples)	1.4 - 5.2	2 - 5.6	-
(6 samples)	-	-	9 - 16.7
Batch Nr 4 (Dounreay)			
Stainless steel - SCOF 2	200 - 570 mean of 10 : 370	54	457 - 919 mean of 4 : 773
Stainless steel - SCOF 3 (60 V)	55 - 550 mean of 10 : 173	18	18 - 63 mean of 21 : 47
Stainless steel - SCOF 3 (93 V)	44 - 200 mean of 10 : 119	23	43 - 69 mean of 5 : 53

(*) Values will be lower on taking account of the Pu-240 content.

2.5.2. Total Content of Activation and Fission Products

The main gamma emitters were determined in completely dissolved hull sections by gamma spectrometry. The results are given in table 2.9.

Table 2.9 : Activation and fission products in :

A - Zircaloy cladding of batch Nr 1 (uCi/g hulls)

Sample (cooling time)	Mn-54	Co-58	Co-60	Zr-95	Ru-106	Sb-125	Cs-134	Cs-137	Ce-144	Eu-152	Eu-154
Nr.3(2245 d)	1.82	-	183	4.3	234	183	84	382	30	-	-
Nr.6(~ 6 y)	-	500	12800	-	700	1300	-	100	-	-	-
Nr.18(2245d)	2.0	-	87	4.5	146	398	106	463	33	-	-
Nr.3 (*)	234	-	406	$1x10^{11}$	$1.5x10^{4}$	828	670	432	$6.7x10^{3}$	-	-

B - Stainless steel cladding of batch Nr 4 (mCi/g hulls)

SCOF 2 (737d)	8.6	0.59	0.57	-	0.89	0.09	-	0.44	0.43	0.09	0.07
SCOF2(*)	44.2	803	0.74	-	3.57	0.15	-	0.46	2.58	0.10	0.08
SCOF 3 (60V) (597 d)	8.1	1.0	0.35	-	-	-	-	0.25	0.22	0.08	0.06
SCOF 3 (60V) (*)	30.5	345	0.43	-	-	-	-	0.26	0.94	0.09	0.07
SCOF 3 (93V) (597-911 d)	7.9	1.0	0.33	-	-	0.06	-	0.10	0.09	0.09	0.06

* Values corrected to time of discharge from reactor

2.5.3. Surface Distribution of Alpha Emitters by Alpha-Spectrometry and Autoradiography

The results of alpha-spectrometric determination are given in Tables 2.10 and 2.11.
Autoradiographs have been made to survey the distribution of alpha activity. A cellulose nitrate film, which is

immune to beta and gamma radiation, was placed in intimate contact with the hull surface. Latent tracks formed by alpha particle can be developed in an etching solution (Fig.2.5.1).

Table 2.10 : Surface activity of KWO hulls (mCi/m^2)

Sample Surface	Pu239+Pu240 (5.15 MeV)	Pu238+Am241 (5.5 MeV)	Cm 244 (5.8 MeV)	Cm 242 (6.1 MeV)	Date of measurement
Batch Nr 1	1.5	2.0	0.94	0.01	14.02.77
2 - Inner	0.05	0.09	0.02	5×10^{-4}	09.02.77
2 - Outer	2.4	3.6	1.3	1×10^{-3}	15.02.77
3 - Inner	0.33	0.17	0.06	5×10^{-4}	19.02.77
3 - Outer	0.74	1.73	0.92	9×10^{-3}	21.02.77
11- Inner	0.26	0.27	0.03	1×10^{-4}	02.03.77
11- Outer	0.96	2.1	0.98	3×10^{-3}	06.03.77
18- Inner					

Table 2.11 : Surface activity of stainless steel hulls (Pu-239 atoms/cm^2)

Surface	Batch sample Nr 4			
	SCOF 1	SCOF 2	SCOF 3 (60 V)	SCOF 3 (93 V)
Inner	3.7×10^{16}	8.3×10^{15}	2.1×10^{15}	6.9×10^{15}
Outer		7.2×10^{16}	1.2×10^{16}	1.0×10^{16}

N.B : By uniform contamination of both surfaces at the level of 1×10^6 atoms 239 Pu/cm^2, one tonne of hulls would contain 27 g of 239 Pu.

2.5.4. Depth Distribution of Alpha Emitters by Alpha Spectrometry

The depth of the emitting nuclide below the surface can be evaluated by the remaining energy of the emerged alpha particle. Fig. 2.5.2 shows some results obtained by this technique. Actinides have penetrated on the inner hull surface to a depth of 1 - 2 microns.

2.5.5. Distribution of Fissile Nuclides by Fission Track Autoradiography

A film of a suitable detector material (e.g. Lexan polycarbonate) is placed in close contact with the surface of the sample, which is mounted in bakelite. After irradiation in a thermal neutron flux ($\sim 2 \times 10^{13}$ n/cm^2/sec), the film is chemically treated to have preferential etching in the regions damaged by fission particles. The tracks can be now observed under optical microscope or with a Quantimet Image Analysing system. A typical example of the distribution of fissile material in Zircaloy hulls is given in Fig. 2.5.3.

Stainless steel hulls were much more heavily contaminated on the outer radius than on the inside but, after cleaning the distribution was reversed (most of outside contamination was removed). Zircaloy hulls were much more contaminated on the inside and the distribution pattern was not changed on cleaning.

2.5.6. Removal of Actinides from Hull Surfaces

Table 2.12 gives results for different reagents used to clean stainless steel and Zircaloy samples in a ultrasonic bath.

Table 2.12 : Removal of actinides from hulls (1 h agitation in ultrasonic bath)

Sample	Reagent/Temperature	Pu-239 (µg/g hulls) removed	Pu-239 (µg/g hulls) left	Percentage removed
Stainless steel				
SCOF 2	3M HNO_3, 22°C	915 - 452	2.5 - 4.1	99
SCOF 3 (60 V)	3M HNO_3, 22°C	20.4- 35.4	1.7 - 9.4	92 - 79
SCOF 3 (60 V)	6M HNO_3, 22°C	34	4.0	90
SCOF 3 (93 V)	3M HNO_3, 22°C	68.2- 62.0	1.3 - 1.2	98
	60°C	41.6	1.3	97
SCOF 2	3M HNO_3 + 0.5 M HF, 22°C	902	<0.25	~100
SCOF 3 (60 V)	3M HNO_3 + 0.5 M HF, 22°C	38.1	<0.25	~100
SCOF 3 (93 V)	3M HNO_3 + 0.5 M HF, 22°C	65.2	<0.25	~100
Zircaloy				
Hull m^3 (Batch Nr 1)	3M HNO_3, 22°C	4.1	5.4	43
" "	3M HNO_3 + 0.5 M HF, 22°C	8.6	8.1	52
" "	0.4 M Ammonium oxalate + 0.16 M Citric acid + 0.05 M H_2O_2, 22°C	3.3	6.9	32

Contamination of both type of hulls (3M HNO_3 at 22°C) may be reduced to the 1-10 µg Pu-239/g hulls range. Using a more aggressive reagent (3M HNO_3 + 0.5 M HF) the stainless steel contamination may be reduced to 0.25 µg Pu-239/g hulls. This reagent did not reduce the Zircaloy contamination below that found for 3M HNO_3. American work (4) has shown that the removal of ZrO_2 (e.g. by treatment with gaseous HF at 600 °C) is necessary to allow a good decontamination of Zircaloy hulls.

2.6. Assessment of the Various Types of Cladding

The Commission has concluded the prospective inventory until 1990 of cladding waste arising in the European Community and their composition. Data have been collected from different Institutions, namely : CEA (France), CEN (Belgium), CNEN (Italy), KFK (Fed. Rep. of Germany) and UKAEA (United Kingdom).

The elaborated data have been published (EUR 5969) (Ref. 5).

2.6.1. Cladding Wastes from Different Reactor Types

Waste amounts and material compositions are summarized in table 2.13. In case of substantial differences in the design of the same reactor type, both values are given.

Table 2.13 : Waste specifications

Reactor	BWR	PWR	SGHWR	AGR	FBR (core)
Waste amounts (Kg/ton H.M.)	400	318-410	282	154	1422 - 1746
Material composition (w/o)					
Zircaloy	80.5	85-72	77	-	-
Stainless steel	9.5	11-22	22.5	95	95 - 100
Harmonic steel	1.3	-	-	-	-
Inconel	8.7	4-6	-	-	-
Various	-	-	0.5	5	5

2.6.2. Waste Arisings in the Community

On the basis of the expected time schedules for the reprocessing capacities in the Member States the cumulative amounts of cladding wastes until 1990 were estimated (Table 2.14).

Table 2.14 : Cumulative arisings of wastes (metric tons).

Country (fuel origin)		1980	1985	1990
France	(LWR)	415	1740	4529
	(FBR)	-	-	- 375
	total	415	1740	4904
United Kingdom				
	(AGR)	-	154	460
	(SGHWR)	-	-	320
	(LWR)	-	-	460
	(FBR)	13	34	55
	total	13	188	1295
Fed.Rep.of Germany				
	(LWR)	75	135	480
Italy	(LWR)	-	-	250
	(FBR)	-	3	10
	total	-	3	260
Belgium	(LWR)	-	75	430
European Comm.				
	(LWR)	490	1950	6140
	(AGR)	-	154	460
	(SGHWR)	-	-	320
	(FBR)	13	38	440
	total	503	2141	7360

These amounts may become substantially lower, as a consequence of further delays in construction and commissioning of new reprocessing plants.

2.6.3. Radioactivity and Decay Heat

The total induced radioactivity and the heat generation of waste components as function of cooling times are given in table 2.15. The neutron exposure rate is 2.9×10^{21} n/cm^2 (thermal flux).

Table 2.15 : Activation (A : KCi/te alloy) and heat generation (H : W/te alloy) at different cooling times.

Alloy	1 y		3 y		10 y		25 y		100 y	
	A	H	A	H	A	H	A	H	A	H
Zircaloy-4	11	77	2.1	27	0.74	11	< 0.1	< 2	-	-
Stainless steel (AISI 321)	437	2300	274	1601	59	572	9.9	75	3.1	0.5
Inconel 718	521	4130	170	1430	73	547	33	78	17	3
Inconel x 750	271	2610	149	1370	79	543	43	80	23	4

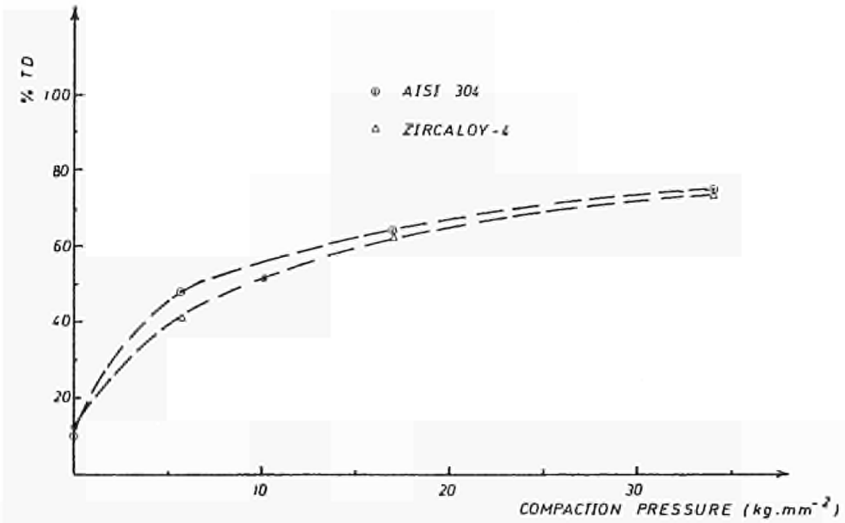


Fig. 2.2.1 : %TH DENSITY Vs. COMPACTION PRESSURE

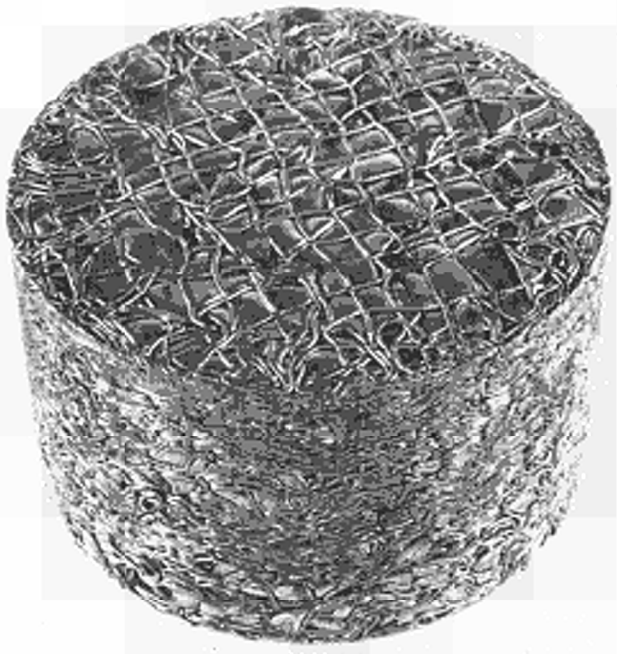


Fig. 2.2.2 : Zircaloy 4 compact to 65 % T.D. (ϕ 150 mm; Ho : 500 mm, Pressure : 167 MPa; densification factor = 5.2)

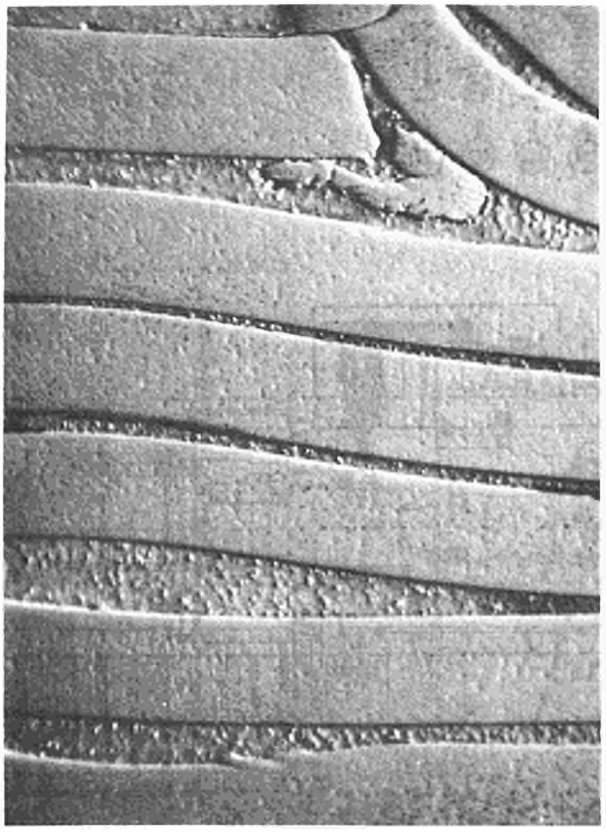
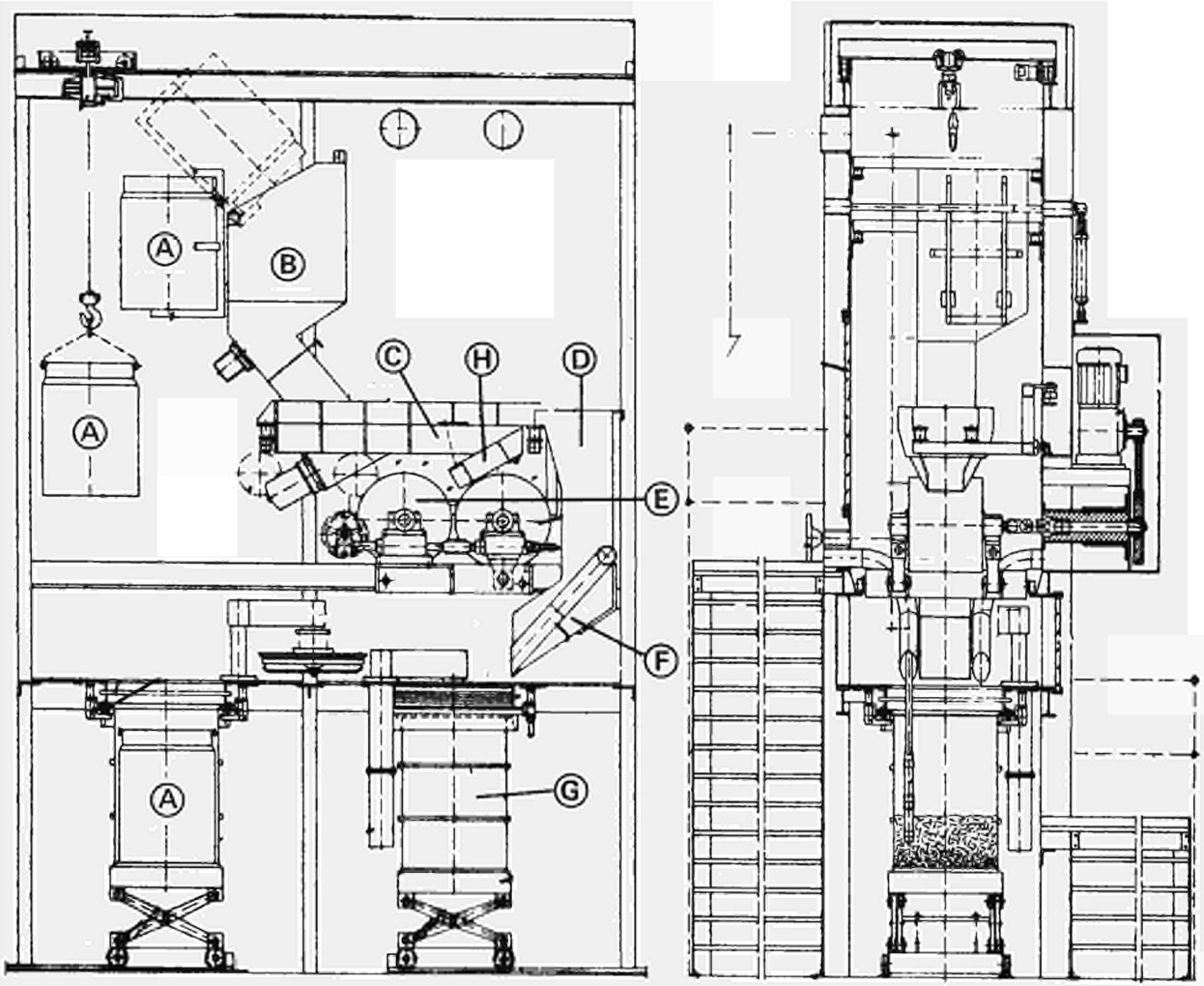


Fig. 2.2.3 : Compacted zircaloy-4 chops (74 % TH) encapsulated by Pb - 1.5 Sb alloy (vacuum casting at 723°K) (Magnification x 20)



Ⓐ= Container of unconditioned cladding waste

Ⓑ= Feeding hopper

Ⓒ= Vibrating runner to separate structural pieces

Ⓓ= Conveying hopper

Ⓔ= Rolling cylinders

Ⓕ= Fluid concrete feeding

Ⓖ= Conditioned waste drum

Ⓗ= Dosing gear

Fig 2.3.1: COMPACTING UNIT

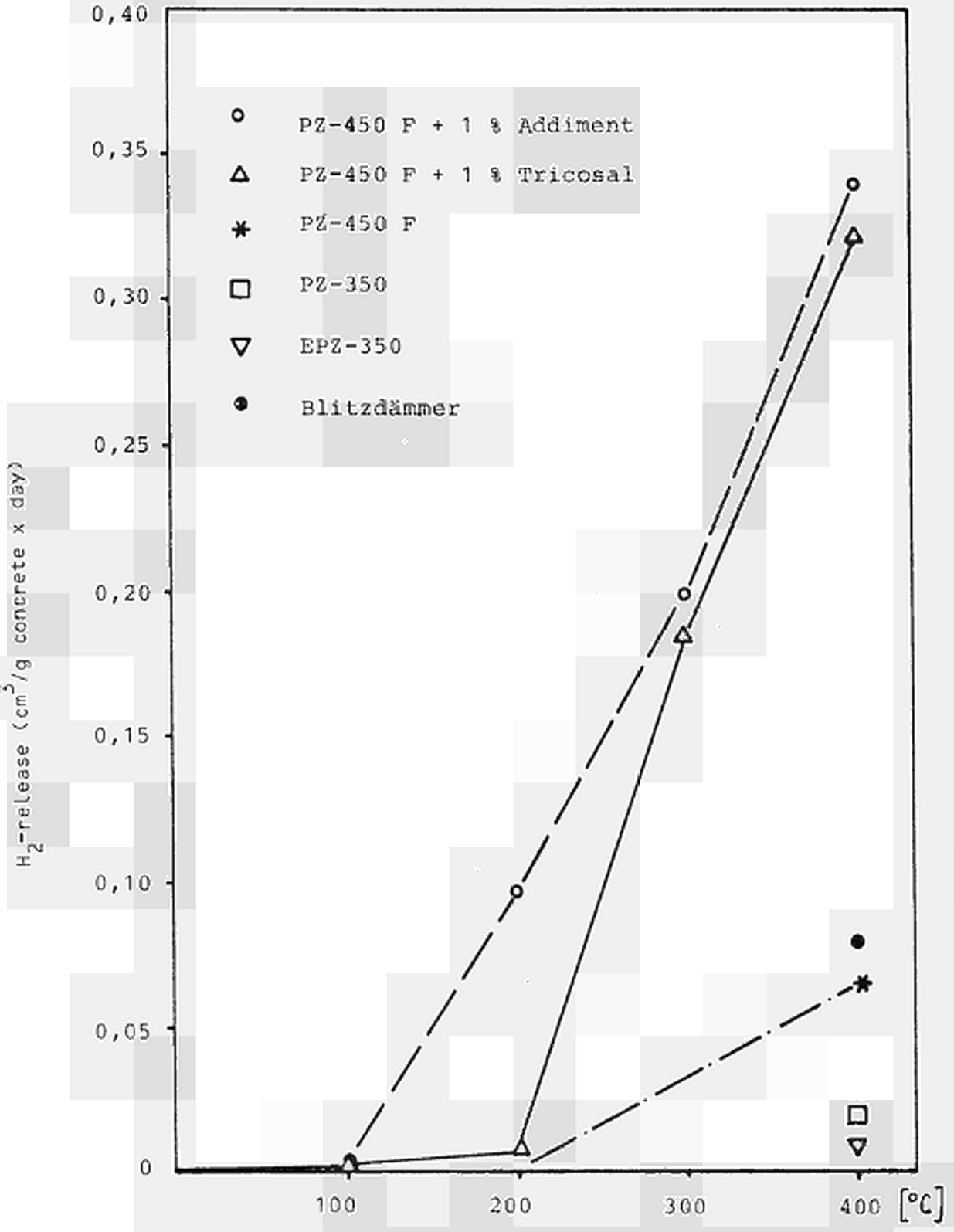


FIG. 2.3.2 : HYDROGEN RELEASE FROM VARIOUS CONCRETE MIXTURES AS FUNCTION OF TEMPERATURE

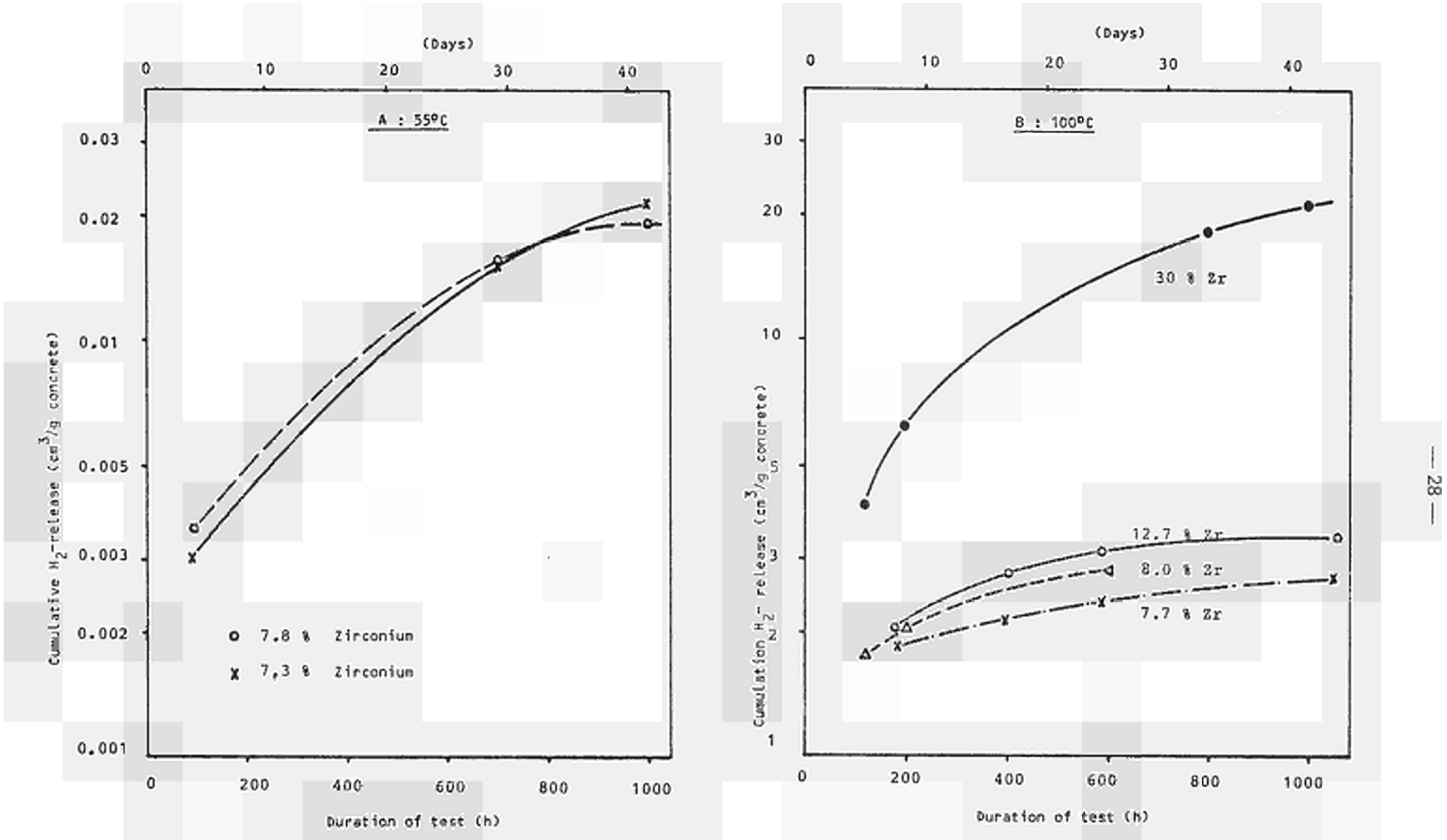


FIG. 2.3.3 : CUMULATIVE HYDROGEN RELEASE FROM CONCRETE (EPZ-350) BY WATER/ZIRCONIUM REACTION AT 55°C (A) AT 100°C (B)

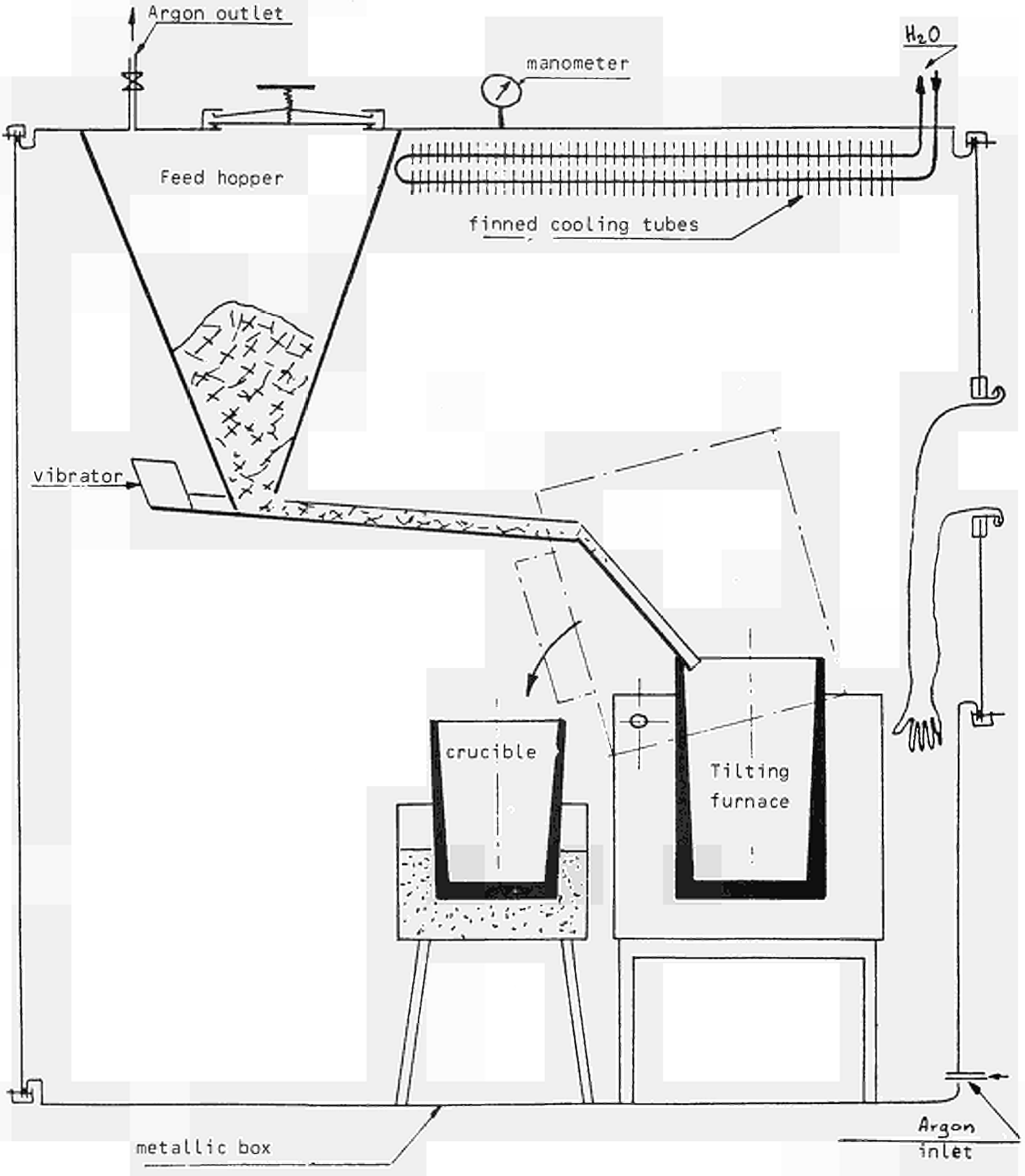


FIG. 2.4.1 : HALF-SCALE MELTING RIG

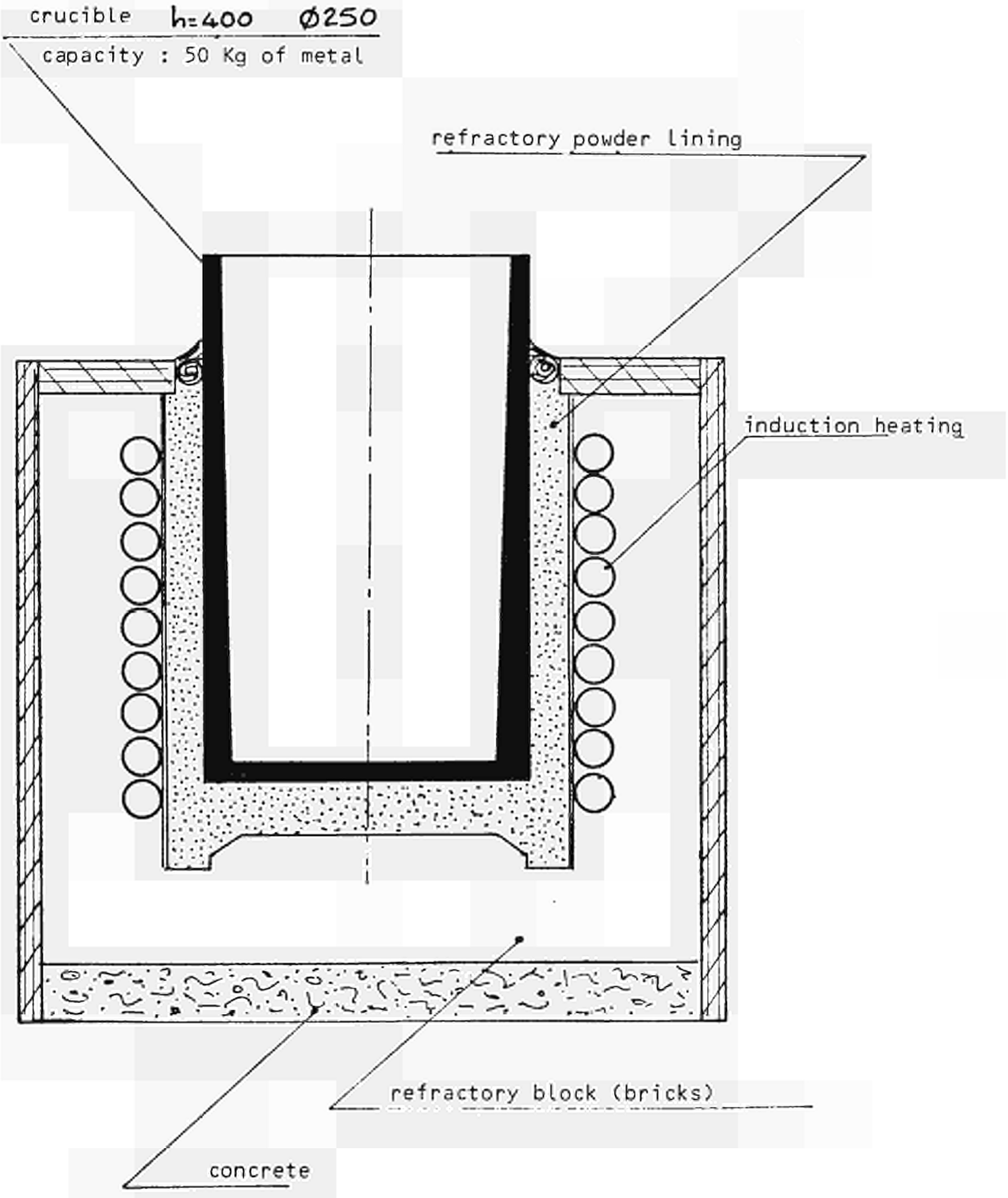


FIG.2.4.2 : MELTING FURNACE (50 KW)

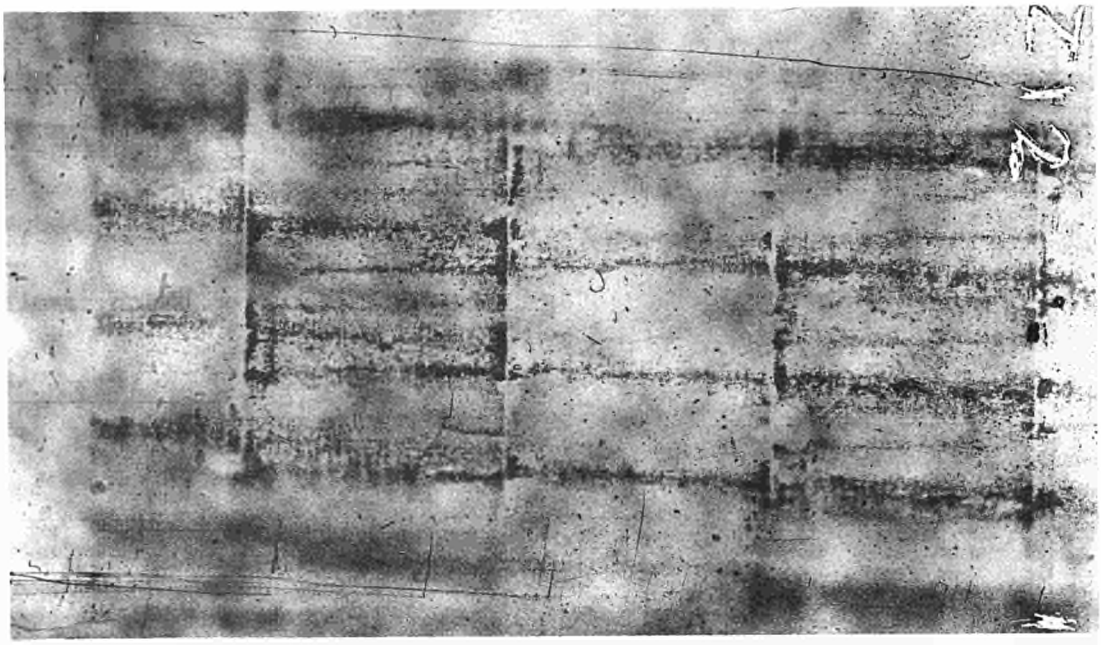
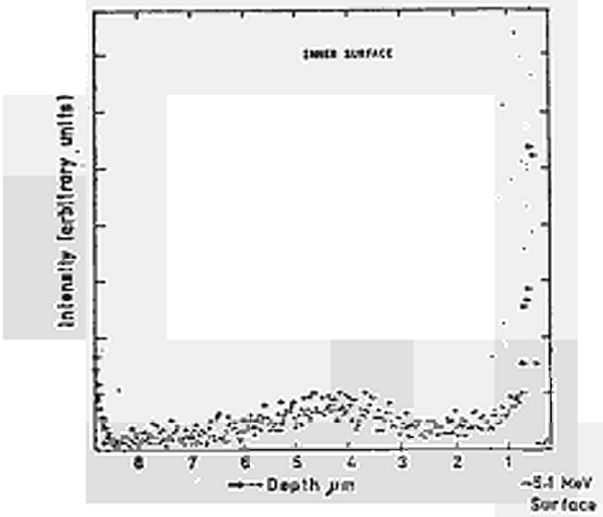
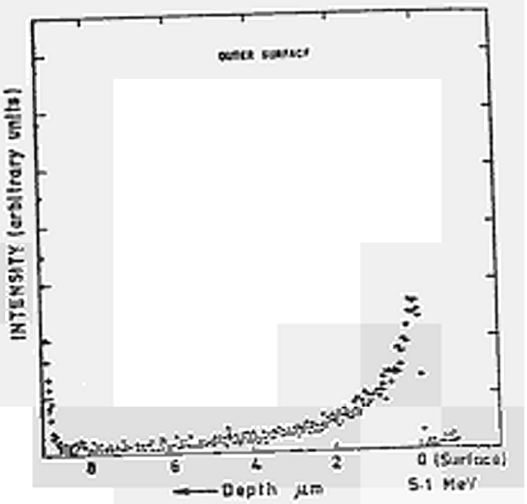
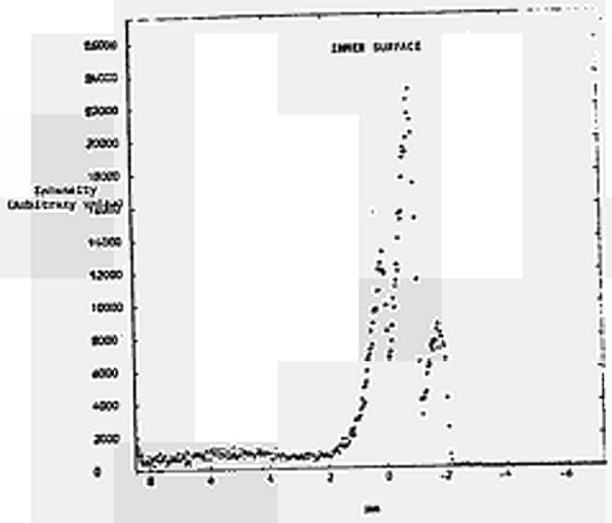


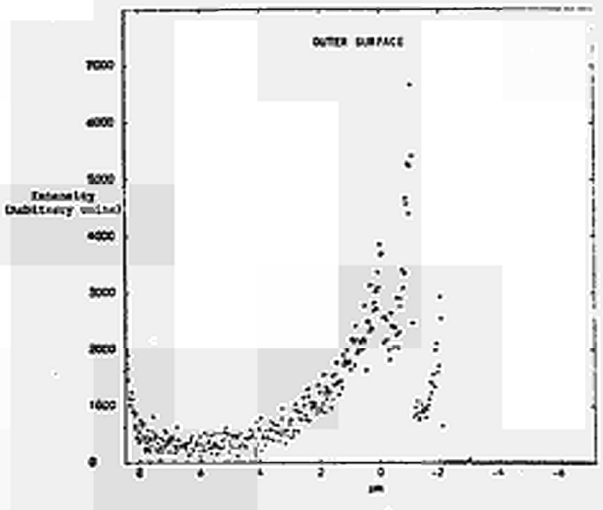
Fig.2.5.1 : Alpha autoradiograph of inside of zircaloy hull (Z 12)
Magnification X3





Stainless steel hulls - SCOF 2 Specimen from batch sample Nr 4





Zircaloy hulls - specimen Nr 3 from batch sample Nr 1

Fig.2.5.2 : DEPTH PROFILE OF Pu-239 ON CLADDING HULLS

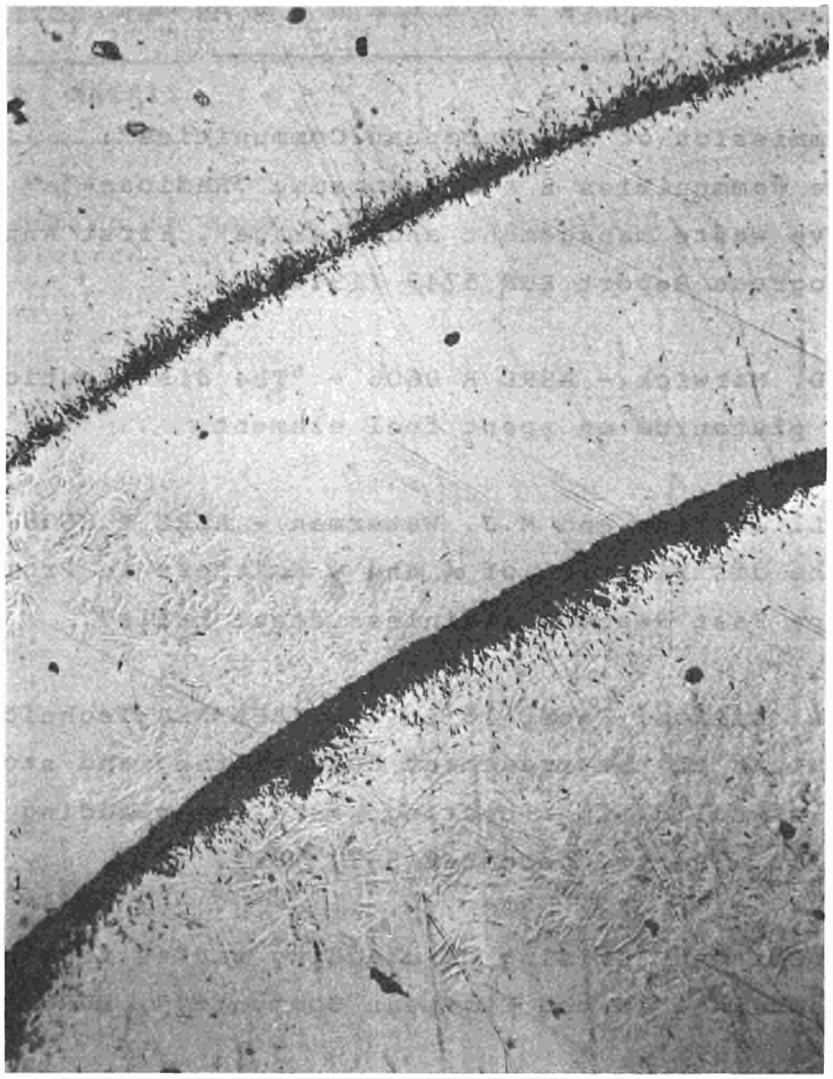


Fig. 2.5.3 : Fissile material distribution on Zircaloy hulls

R E F E R E N C E S

(1) Commission of the European Communities : The Communities R & D Programme "Radioactive waste management and storage", First Annual Progress Report EUR 5749 (1977).

(2) A.D. Marwick - AERE R 8606 - "The distribution of plutonium on spent fuel element.

(3) J.L. Jenkins and M.J. Waterman - AERE R 8608 - "The distribution of α and γ emitters in Prototype Fast Reactor stainless steel hulls".

(4) R.L. Dillon, Paper 16 in the IAEA/NEA Technical Seminar on the treatment conditioning and storage of solid alpha-bearing waste and cladding hulls - Paris, December 5-7, 1977.

(5) G. Cottone "Arising of cladding wastes from nuclear fuel in the European Community", EUR 5969 (1978).

3 IMMOBILIZATION OF FISSION PRODUCT CALCES

A METALLIC MATRIX

There is nothing to report since this action has been deferred.

4 TREATMENT OF Pu-CONTAMINATED COMBUSTIBLE WASTES

4.1. General Aspects and Tendencies

Plutonium contaminated wastes or more generally α-wastes mainly arise from such operations at the back end of the fuel cycle as the extraction cycles of reprocessing, the reconversion of recycled fuel and the manufacture of MOX fuel elements. A substantial part of this Pu-contaminated waste is combustible e.g. the gloves from glove-boxes, used filters, packing material and protective clothing.

The Community programme promotes five incineration studies and one specific investigation on Pu recovery.

The broad aim of these projects is to :

- reduce the volume of waste;
- obtain waste products, which are compatible with available conditioning and disposal methods;
- separate the actinides from other residues;
- recover plutonium where practicable.

During 1977, most of the new actions were placed with studies totally or largely devoted to the recovery of Pu.
Among the most difficult problems encountered by all investigators was to ensure satisfactory off-gas cleaning. Other problems not yet solved in many cases concern the construction materials and the process control.

4.2. High Temperature Incineration (CEN/SCK-B)

4.2.1. Objectives

The objective of this contract is to develop the high temperature incineration technique for the treatment of plutonium (Pu) contaminated waste. Two aims are being pursued :

a) Incineration of low Pu-contaminated waste to produce a highly leach resistant product sui-

table for long term storage and ultimate disposal by means of a modified commercial waste incinerator (FLK60, capacity 100-150 kg/h), (Ref. 1);

b) Incineration of heavily Pu-contaminated waste, with the aim to produce a readily soluble product in order to promote maximum Pu-recovery by means of a reduced scale pilot plant in development (capacity 10 kg/h).

4.2.2. Design and Construction Work

4.2.2.1. Installation and Testing of the Off-Gas Purification System

The installation of the off-gas purification system to the FLK60 plant was completed consisting of :

- 2 heat exchangers;
- 2 panel bed filters;
- scrubber unit;
- 2 absolute filters;
- gas control system;
- ventilation system;
- stack.

The system has been tested in parts and as a whole and repeatedly modified. Apart from the panel bed filters, whose retention capacity was found to be insufficient, the off-gas system satisfactorily performed. But neither PVC nor rubber have yet been treated. Preliminary tests on the efficiency of the scrubber unit for trapping gaseous HCL have been carried out by injection of pure HCL. 95 % of the HCL could be trapped and it is believed that this can be substantially improved by adding Na OH. Extensive particle size distribution measurements of fly ashes are being carried out in situ. A safety report for the off-gas system is in preparation.

4.2.2.2. Optimization of the FLK60 Plant and its Adaptation for Pu-Contaminated Waste (Fig. 4.2.1)

The waste feed device has been modified and successfully tested.

The FLK60 plant has started operation with traces of β,γ -activity in January 1978.

4.2.2.3. Design and Construction of the Alpha Containment and the Waste Pre-Treatment Facilities

A layout of the alpha containment is given in Fig. 4.2.2.

During 1977 parts of the waste pre-treatment equipment have been constructed and tested. The design for the alpha containment was completed and construction work will start 1978.

4.2.2.4. Installation of an Active Granules Characterization Laboratory

Parallel to the inactive laboratory an active one is being installed to handle medium active material. The following test equipment for the granules characterization will be included :

- equipment for manufacture and preparation of samples;
- instrument for melting point measurements;
- a furnace to simulate the melting process;
- a furnace to produce ash samples;
- a leaching device for Pu-extraction tests.

4.2.2.5. Design of the Pilot Plant (Fig. 4.2.3.)

The general design of the pilot plant is completed and a tender has been sent out for the construction of the plant during 1978. A criticality study is being carried out.

4.2.3. Experimental Work

4.2.3.1. Experimental Operation of the FLK60 Plant

126 inactive but full scale experiments with a variety of simulated waste compositions have been carried out with the FLK 60 plant. Each experiment amounted to approximately 600 kg of treated waste. The composition of granules for various residence times (table 4.2.2) show no hold-backs of elements in the incinerator.

4.2.3.2. Leaching Tests

The incremental leaching rate has been determined according to IAEA specifications (Ref. 2) of the ash granules from various experiments. The average leaching rates for Fe^{59}, Cr^{51} and Co^{60} are given in table 4.2.3.

4.2.3.3. Structural Tests

The compositions of the original waste and the respective incineration ash investigated are given in tables 4.2.4 and 4.2.5. The reaction melting temperature of the mixture was lying between 1400 °C and 1460 °C. The melting point of the granules was 1360 °C ± 10 °C. The residual carbon content was decreased from 0,42 % to 0,05 %. The "air" density of the obtained granules was $\simeq$ 3,4 g/cm^3, "tap" density $\simeq$ 1,7 g/cm^3.

Microprobe analysis yielded to the conclusions :

- that the iron present is alloyed to all the other components (especially to Cr) except to NiS;
- that sulphur is found in the $BaSO_4$ matrix and in concentrated form in the Ni-phase.

Some scanning electron microscopes have been made showing in one case a pronounced glassy phase. A statistical analysis of granules respectively reacted and non-reacted materials is given in table 4.2.6.

4.2.3.4. Laboratory Experiments

Laboratory tests have been carried out to study the influence of waste components on the melting point of the granules, the volatility of some elements during the incineration and melting process and the solubility of some components on the final granules. The volatile components of ash granules was measured and amounted in some cases up to 4,5 % at 850 °C. Samples of simulated waste constituents and compositions were investigated. Incineration - and - melting point measurements with a hot-stage microscope showed the following relations :

- The incineration point of the green waste increases with the increase of Fe content;
- The melting point of the melted waste increa-

ses with the increase of Fe content and decreases with the increase of SiO_2 and CaO contents.

Cylindrical pellets traces with Ru^{106} and C_s^{137} have been heated in a pre-heated oven at 1400 °C for 15 minutes. The rate of volatility has been determined by the loss of the two isotopes counting the remaining activity. Hence the volatility behaviour has been investigated in relation to the concentration of certain constituents, of which the following tendencies have been observed :

- decrease of C_s volatility with an increase of SiO_2 + Fe_2O_3 concentration as well as with an increase of acid components;
- maximum C_s volatility at ~45 % of alkaline content (decreasing above and below 45 %);
- maximum Ru volatility at ~40 % of SiO_2 + Fe_2O_3 concentration (decreasing above and below 40 %);
- minimum Ru volatility at ~35 % of alkaline content (increasing above and below 35 %).

Samples doped with C_s^{137} have been leached according to the specifications mentioned in chapter 4.2.2.1, the results of which are plotted in Fig. 424.

4.2.3.5. Study of Waste Compositions and Additives with the Aim of Highly Soluble Incineration Ashes

Investigations have been carried out on waste compositions and additives. The composition defined as the most waste mixture to start the pilot plant experiments with, in accordance with the experience gained from the FLK 60 operation, is given in table 4.2.7. A statistical composition of the expected ash product is given in table 4.2.8. During the first tests plutonium will be substituted by cerium. According to a literature survey a mixture of Na_2CO_3-Na_2SO_4-NaCL as additives seems to be most promising.

Up to 99 % of the plutonium can be retained in the fluid ash-salt mixture. The melting point of the mixtures is kept low (~800 °C) by the additives which is promoting a better soluble product for the Pu-extraction tests.

4.2.4. Interim Conclusions

The experimental operation of the FLK 60 plant has demonstrated that, after some modifications and apart from the failure of the panel bed filters, trouble free incineration at temperatures up to 1500 °C can be carried out. The leaching results on Cr^{51} indicate a generally good retention of the radioactive isotopes. The laboratory techniques for the simulation and characterization of granules are promising for use under Pu-containing conditions. The main actions of the programme are about one year behind the original schedule.

4.2.5. Planned Activities

As indicated in the time schedule table 4.2.1, 1978 will be devoted to :

- the experimental operation of the FLK 60 plant with traces of β, γ-activity and in conjunction;
- the experimental research of the influence of additives on ash quality and off-gas (aim = high solubility);
- the completion of the active "granules characterization laboratory";
- the repair of the sludge drying system;
- the construction of the alpha containment and the installation of the waste pre-treatment facilities and
- the construction of the pilot pilot plant (capacity 10 kg/h).

4.2.6. Complementary Action

The research programme has been expanded starting July 1977 by the complementary action "densification of the ash granules derived by incineration". The objective is to develop a technique suitable for the final conditioning of the ash residues originating from low plutonium contaminated waste. The method investigated is to compress the ash granules at high pressure (up to 100 kg/mm^2) and high temperature ($\sim$800 °C) in order to achieve a supplementary volume reduction of approximately 50 % and an immobilized end product. An experimental programme to

prove the feasibility of the process and to optimize the process parameters - pressure, temperature, duration of compression and obtained densification - is carried out with a work-shop press during 1977/78 on inactive samples. The test samples will be examined with respect to leachability and microstructure during 1978/79.

4.2.7. References

(1) First Annual Progress Report, contract EUR-017-76-7 WASB, N. Van de Voorde et. al.

(2) Atomic Energy Review, Vol. 9 N° 1, E.D. Hespe, IAEA Vienna 1971.

Table 4.2.1 : Timetable for Incineration Programme

Programme Item	1976	1977	1978	1979
1. FLK60 Plant				
1.1 Operation with inactive waste (progress report 23.2.77)				
1.2 Research of influence of waste composition on asa quality, 126 experiments and evaluation (aim = high insolubility)				
1.3 Adaption and optimization for Pu-contaminated waste (waste supply, ash retrieval)				
1.4 Construction and installation of off-gas purification system				
1.5 Operation with traces of α -activity				
1.6 Research of influence of additives on ash quality and off-gas (aim = high solubility)				
1.7 Operation with low Pu-contaminated waste				
2. Other Facilities				
2.1 Installation of active "granules characterization laboratory"				
2.2 Construction of sludge drying system (and repair)				
2.3 Design of α -containment and waste pretreatment facilities				
2.4 Construction of α -containment and waste pretreatment facilities				
3. Pilot Plant				
3.1 Design study				
3.2 Construction				
3.3 Operation with Pu-waste				
3.4 Pu-recovery tests				

Table 4.2.2 : The composition of granules at various residence times in the reactor.

Element	Sample taken at various smelting hours (%)						
	1st.	4th	5th	10th	15th	17th	19th
Al	3.6	2.8	4.6	4.2	4.4	5.1	5.5
Ba	2.4	3.2	3.5	2.1	1.7	3.3	1.9
Ca	0.51	0.76	0.54	0.51	0.54	1.05	0.7
Cr	0.29	0.52	0.14	0.18	0.23	0.28	0.14
Fe	3.6	3.6	4.3	3.5	2.7	3.4	2.6
Mg	0.08	0.12	0.08	0.07	0.04	0.1	0.05
Mo	0.04	0.03	0.03	0.02	0.03	0.02	0.02
Na	3.9	2.9	4.2	3.9	3.5	2.8	3.3
Ni	0.57	0.42	0.61	0.36	0.40	0.30	0.25
Si	31.4	33.6	27.9	31.4	33.4	33.4	32.7
Sr	0.04	0.01	0.06	0.04	0.03	0.02	0.05
Ti	0.45	0.32	0.44	0.04	0.03	0.72	0.43
Zn	0.28	< 0.1	0.44	0.32	0.41	traces	0.41
Cu	6.6	4.5	9	6.4	4	4.1	4.1
Mn	0.11	0.13	0.12	0.13	0.13	0.15	0.13

Table 4.2.3 : Leaching factor after 236 days - Leaching in demineralised water

Sample	Isotope	$g.cm^{-2}.day^{-1}$ L x D	$cm.day^{-1}$ L
76/33/04	Cr^{51}	$3,09 \times 10^{-10}$	$9,08.10^{-11}$
	Fe^{59}	$4,35 \times 10^{-11}$	$1,28.10^{-11}$
	Co^{60}	$1,62 \times 10^{-10}$	$4,78.10^{-11}$
76/33/06	Cr^{51}	$1,40 \times 10^{-10}$	$4,19.10^{-11}$
	Fe^{59}	$4,34 \times 10^{-11}$	$1,30.10^{-11}$
	Co^{60}	$8,52 \times 10^{-9}$	$2,55.10^{-9}$

$$L = \frac{\pi m^2 V^2}{4F^2} \ .cm.day^{-1}$$

D = density.

Table 4.2.4 : Composition of artificial waste (code : US 2/5).

Components	%
Combustible :	
paper	28.3
plastics (P.E.)	14.3
ion exchangers	+ 9.5
	52.1
Metals	9.5
Sand	6.7
Glass	1.9
Na_2CO_3	1.9
Water	17.1
Sludge (x)	10.8

(x) Sludge composition :

$BaSO_4$	:	43.29
Fe_2O_3	:	35.11
Al_2O_3	:	6.25
SiO_2	:	5.00
$BaCO_3$	:	4.81
CuO	:	2.88
MgO	:	0.76
MnO_2	:	0.76
$Ca(OH)_2$	:	0.62
TiO_2	:	0.48

Table 4.2.5 : Chemical composition of the granulate resulting from the incineration of waste.
Type code : US 2/5

% of the component comprised in a granulate taken after :				
	1 hour	2 hours	4 hours	6 hours
SiO_2	66.9	63.0	67.3	87.2
Fe_2O_3	8.1	6.5	6.9	3.2
Al_2O_3	9.9	11.5	10.2	2.0
Na_2O	5.0	7.8	5.8	2.9
BaO	2.3	2.4	2.1	0.89
MgO	0.52	0.57	0.38	0.22
MnO	0.40	0.46	0.40	0.20
Cr_2O_3	0.87	0.62	0.93	0.24
Mo_3O_4	0.12	0.15	0.15	0.03
CuO	3.10	3.98	2.97	2.47
TiO_2	1.29	1.23	0.83	0.23
NiO	0.70	0.66	1.41	0.14
$SrCO_3$	0.09	0.13	0.07	0.02
CaO	0.67	0.89	0.46	0.12
C	0.26	0.43	0.06	0.05

Table 4.2.6 : Statistical analysis of granulate on reacted and non-reacted material.

	Granules fraction (w%)			Non-reacted metal (w%)			Non-burnt paper and plastics (w%)		
Fraction - mm	max.	min.	mean	max.	min.	mean	max.	min.	mean
> 20	16,2	0,5	4,28	2,48	0,0	0,15	2,47	0,0	0,20
> 10	42,1	2,0	17,46	0,54	0,0	0,20	0,99	0,0	0,12
> 5	33,1	7,9	21,96	0,58	0,0	0,12	0,32	0,0	0,04
> 2	53,9	16,1	31,13	0,25	0,0	0,02	0,22	0,0	0,03
> 1	30,1	4,4	15,74	-	-	-	0,08	0,0	0,01
> 0,5	13,8	2,8	4,82	-	-	-	0,08	0,0	0,01
> 0,25	3,7	0,5	2,12	-	-	-	-	-	-
> 0,1	9,1	0,1	1,29	-	-	-	-	-	-
< 0,1	8,3	0,3	0,96	-	-	-	-	-	-

Table 4.2.7 : Composition of alpha-contaminated waste with a high PuO_2-content (> 10 g Pu/28 l)

Component	Vol %	W %
plastics	34	24
paper-hardboard	30	20
rubber	20	25
metals	10	12
cellulose	5	2
wood	1	6
moisture in components	-	10
Pu content	> 10 g/28 l	0,5

Table 4.2.8 : Estimation of the ash composition of the burnable parts of alpha-contaminated wastes with high PuO_2-content.

Component	% incinerator ash AB	% ash from the incin. of rubber AR	% $\frac{AB}{AR} = \frac{1}{20}$
BaO	0,35	-	0,02
CdO	0,02	-	-
B_2O_3	0,81	-	0,04
MnO	0,65	1,3	1,27
Fe_2O_3	7,94	13,3	13,04
MgO	2,87	6,7	6,52
SiO_2	48,38	20	21,35
PbO	1,44	-	0,07
Al_2O_3	6,00	26,7	25,71
CuO	2,82	1,3	1,37
TiO_2	8,41	1,3	1,64
CaO	10,32	6,7	6,87
SnO_2	0,10	-	-
Cr_2O_3	1,93	-	0,09
ZnO	1,77	20	19,13
SrO	0,09	1,3	1,24
MoO_3	0,11	-	0,01
K_2O	1,58	-	0,08
Na_2O	0,18	-	0,01
ZrO_2	3,59	-	0,17
P_2O_5	0,52	1,3	1,26

AB : ashes from burnable parts except rubber;
AR : ashes from rubber.

4.3. Incineration in Molten Salts (Agip Nucleare)

4.3.1. Objective and scope

The Molten Salt incineration technique permits waste incineration and plutonium separation a continuous process by destroying the combustible waste in a molten salt bath with controlled air as oxidant, the plutonium remaining dissolved in the molten salts after addition of sulphuric acid (Fig. 4.3.1).

A second step would recover the plutonium from the salt.

By filtration all the non consumed and incombustible waste would be separated from the plutonium (and other heavy metals) dissolved in the fused salt.

The technique studied by Agip Nucleare, unlike Atomics International's molten salt process, will use alkaline instead of carbonates for the Pu-separation. The chemical reactions following the oxidization of the waste as well as the dissolving and precipitating of the plutonium have to be investigated in detail before proceding to the design and construction of a pilot plant.

The first part of the programme will investigate the combustion of the waste (without Pu) and, separately, the dissolution of ashes and uranium and the recovery of uranium.

The second part of the programme consists of the operation of a bench scale rig which can, on a small scale, perform the combustion process with Pu contaminated waste. The Pu recovery, presently tested in a separate unit, will be incorporated into the bench scale rig.

In addition to this Pu-bearing test facility, construction and operation of small pilot plant for the treatment of uranium contaminated waste is envisaged.

4.3.2. Results from Combustion Tests

4.3.2.1. The Waste

The composition of the simulated waste in wt % used in the tests is :

Plastic	PVC	40
	PE	20
Rubber		24
Paper		6
Rags		5
Wool		5

The bulk density of the mixture is 190 kg/m^3, its heating value 30.1 x 10^6 - 31.4 x 10^6 J/kg and its ash content 3.34 %.

4.3.2.2. The Molten Salts

The initial composition of the molten salt bath is (Mol %) :

Li_2SO_4 78 %

Kr SO_2 8,5 %

Na_2SO_4 13,5 %

This eutectic mixture melts at 512 °C.

The combustion rate of the bench scale test rig is 15 - 20 g/h with a 1/3 air-nitrogen flow of 0.4 m^3/h.

The molten salt charge of the experimental incinerator is 5 kg.

After processing about 1 - 2.5 kg of inactive waste, the melt is filtered through a 100 µ sintered SS filter and recycled.

4.3.2.3. The Incineration Residues

The reduction factors derived for the total filter residue (ash + salt) range from :

3.3 - 10.5 for the weight reduction
and 30 - 114 for the volume reduction factor.

In fact, the weight reduction based on the waste/ash ratio, i.e. after washing the filter residue, is twice to four times higher.

4.3.2.4. Combustion Gases

After the combustion reaction, at temperatures around 550-600 °C, the off-gases (N_2, O_2, CO_2, CO, H_2, H_2O = steam, hydrocarbons as well as Cl_2 and HCL from the PVC) are burned in a post-

combustion chamber, where the organic matter is completely destroyed at temperatures around 1000 °C.

The small amounts of HCL and chlorine gas left in the off-gas after the post combustion are sufficiently washed out but not completely in caustic solution.

The amount of particular matter ($\sim$20 mg/m^3) remaining in the off-gas after caustic scrubbing is retained in an absolute filter.

4.3.2.5. Pu Recovery (Simulation)

The recovery technique for plutonium has been simulated with uranium, added to molten salt containing incineration ashes from combustion tests.

The separation and recovery tests are performed in a quartz vessel. By adding 50 cm^3 of 100 % H_2SO_4 for each kg of molten salt, the uranium oxide and all the ashes with exception of silica is dissolved.

The uranium oxide (purity = 80 - 90 %) is then separated to Ca 90 % from the molten salt bath by electrolysis. As the ashes mainly consist of metallic impurities (s. tab. 2) some of the latter (Fe, Al, Zn) will also be partially deposited on the electrode, unless removed by pre-treatment.

An attempt to precipitate the Al, Fe, Zn and Cr by thermal decomposition of the respective soluble sulphates to insoluble oxides during a prolonged ($>$24 h) heating at 600-650 °C succeeded to remove only about half of these impurities. This result does not justify the inconvenience of the added process steps.

4.3.2.6. Recycling the Molten Salt

The components of the ashes remaining in the molten salt after the electrolysis can be eliminated either by thermal decomposition or by precipitation with Na_2CO_3.

The results of the thermal pre-treatment, electrolysis separation and post-electrolytic cleaning of the molten salt with Na_2CO_3 are presented in Table 4.

4.3.2.7. Materials

The bench-scale test rig (Fig. 4.3.2) is mainly constructed of refractory steel AISI 310 S, which resists chemicals as molten sulphates at temperatures up to 1000 °C. Nevertheless it is attacked by the chlorine and hydrochloric acid in the off-gas at high temperature.

A major material problem is the dissolving of the ashes, when 100 % pure H_2SO_4 and alkaline bisulphates are added to the molten sulphates at a temperature of 500-600 °C. None of the metallic materials tested except platinum resisted under these conditions.

Quartz and alumina vessels proved quite suitable, but the materials are very brittle.

A refractory crucible with an 0,5 mm alumina coating applied by plasma did not overcome the problem due to the microporosity of the coating.

4.3.3. Forward Programme

During 1978 the bench scale incineration rig and the Pu-separation test equipment will be installed and operated in an alpha containment at Ispra.

A small pilot plant for incinerating uranium bearing waste and recovery of uranium will also be designed and constructed.

4.4. Acid Digestion Process (KFK, FRG)

4.4.1. Objectives

KFK is continuing the work, started by Nukem, on the acid digestion process (flow sheet given in Fig. 4.4.1) in order to develop the process to a stage which permits construction and operation of an active pilot plant.

The items to be investigated are :

- Isolation and recovery of plutonium from process residues;
- Recovery and recycling of sulfuric and nitric acids from off-gases;
- Transformation of remaining nitric components of the off-gas in inert gases;

- Optimization of the combustion process.

The process consists in breaking up the organic components of the burnable solid waste by using sulfuric acid. The subsequent oxidation is achieved by addition of nitric acid. The reaction products are gases and inorganic residues.

All common burnable solid wastes of the nuclear cycle are rapidly consumed at 250 °C.

4.4.2. Scope of the Programme

The research programme which started on 1 July 1977 and will end on 31 December 1979 will investigate the following subjects :

a) Kinetics and optimization of the sulphuric-nitric acid system (4.4.2.1);

b) Separation of the acid from the off-gas wash solution (4.4.2.2);

c) Conversion of unabsorbable acid gases into exhaustible off-gases (4.4.2.3);

d) Separation of the plutonium components (4.4.2.4);

e) Plutonium recovery from the plutonium compounds (4.4.2.5).

4.4.2.1. Combustion Process

To investigate the kinetics in the system H_2SO_4-HNO_3 for the treatment of simulated (inactive) solid waste and to optimize the combustion process, a laboratory scale test plant (ILONA)+ with a capacity of 1,5 kg/h at continuous feed will be designed and constructed during 1978. The plant will be operated during 1979 to determine conditions for the minimum acid consumption, optimum reaction temperature and dimensions of the reactor vessel as well as dosing and location of the HNO_3 addition.
A standard waste composition for the various process steps (table 4.4.1), which will be the basis for all tests, has been agreed with Eurochemic.

+whose mass-flow-sheet is given : Fig. 4.4.2.

Table 4.4.1 : Composition of standard waste

Component	Parts in standard waste (wt. %)
Cellulose	15 - 40
Neoprene	20 - 40
Polyethylen	20
PVC	20 - 25

4.4.2.2. Recycling of Reaction Acids from the Off-Gas System

In order to reduce the secondary waste from the off-gas[+] system rectifying columns for the distillative separation of sulfuric-nitric and hydrochloric acid from the off-gas wash solution are being developed. A bench scale rig, designed for inactive trials, with a capacity of 0,7 l/h will be connected through a bypass to the off-gas system of the combustion plant (chapter 4.4.2.1).

Experiments have been carried out with a test mock-up (Fig. 4.4.3) to determine the concentration profiles for H_2SO_4 and HNO_3 of the distillation devices at various test conditions.

The acid recycling system will consist of two distillation steps : step 1 separation of H_2SO_4 from HNO_3 and HCL and step 2 - HNO_3 separation from HCL. The concentration profiles anticipated are : for H_2SO_4 from 15 % to 80-90 % and for HNO_3 from 20 % to 50-60 %.

4.4.2.3. Conversion of non Condensable Acid Gases to Exhaustible Off-Gases

In order to remove the remaining acidic off-gases (NO_x and NOC_l) the reduction by formic acid will be investigated during 1978. The

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[+] essentially CO_2, NO_x, SO_2, HC_l and water vapour.

thus formed hydrochloric acid will be converted to salt. Another bench scale rig will be constructed and connected with the off-gas system of the combustion plant (chapter 4.4.2.1). Suitable construction materials for the off-gas system will be examined and the material data of the reaction determined.

4.4.2.4. Separation of Plutonium Concentrates from the Acid Solution

For the demonstration of the separation process plutonium concentrates will be separated from the acid by means of a centrifuge (capacity 1.5 kg/h). This plant item will be constructed during 1978 and operated with inactive residues during 1979. The objective of these experiments is to optimize the separation process (size of centrifuge, number of revolutions, effect of precipitants). The composition of the plutonium bearing concentrate and the volume reduction waste/residue will be determined and a criticality study will be carried out.

4.4.2.5. Isolation of Plutonium from the Plutonium Concentrate

An alpha box-line with the appropriate equipments has been installed during 1977 at the site of Eurochemic Mol for the Pu-recovery experiments. A small combustion unit (Fig. 4.4.4) and a small centrifuge (capacity 1 kg/h) have been installed in glove boxes for the preparation of combustion residue and for the separation of the plutonium concentrate.

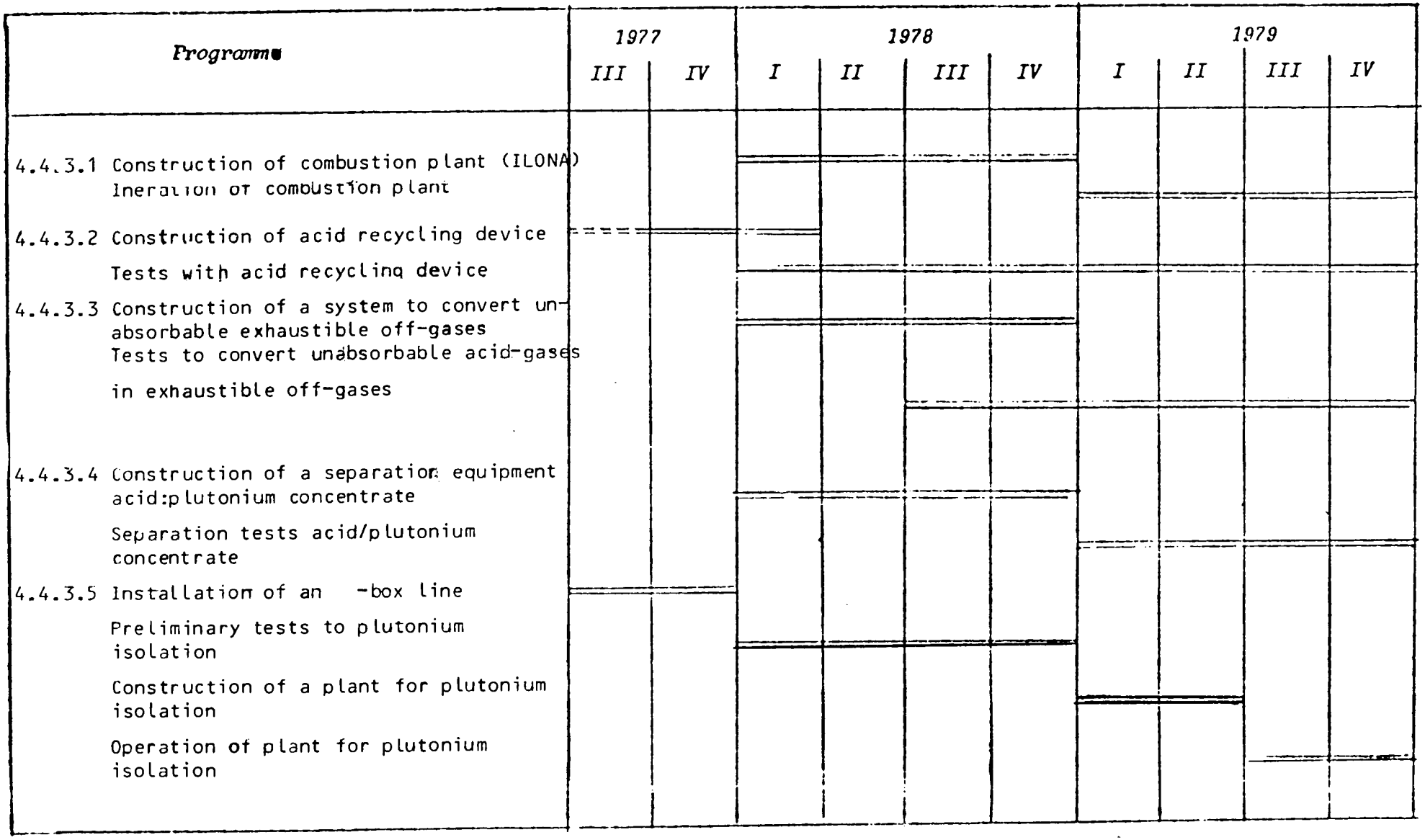
An experimental plant for the plutonium isolation, treating plutonium in amounts of grams (corresponding a waste quantity of 0,5 kg) will be constructed during the first half of 1979 and installed in a glove box. This test unit will be operated with inactive residues from the wet combustion plant (chapter 4.4.2.1), subsequently doped with plutonium, and with plutonium contaminated waste from Eurochemic (2 to 3 g Pu/kg waste). The objective of these experiments is to optimize the plutonium isolation process and examine the qualification for the conversion of the extract into plutonium nitrate. Various means of extraction will be tested and the res-

pective extraction rate, the co-precipitation of actinides and the quantities of plutonium remaining in the acid and the residue will be determined.

4.4.3. Status of Advance

As indicated in table 4.4.2 the first six months of the research programme have been exclusively devoted to the construction of experimental equipments. No experimental results have been achieved so far.

Table 4.4.2 : Timetable for the research programme "wet combustion of plutonium contaminated waste".



4.5. Process for the Pyrolysis of Plutonium Contaminated Combustible Solid Waste (UKAEA HARWELL)

4.5.1. Objectives

The broad objective of this programme is to provide data for an engineering appraisal of the pyrolysis process as applied to plutonium contaminated combustible solid waste and for the design of an inactive pilot plant.

4.5.2. Research Programme

4.5.2.1. Scope of the Programme

The proposed programme will examine :

- The performance parameters of a pyrolysis reactor;
- The off-gas treatment parameters;
- The handling and treatment of pyrolysis residues (char);
- General plant design and operation.

The construction and operation of an inactive pilot plant is envisaged following the completion of the present programme.

4.5.2.2. Reactor Performance Parameters

The effect of the important parameters :

- Temperature;
- Heat transfer rate;
- Feed composition and
- Size

upon the pyrolysis rate and upon the quantity of solid and gaseous product will be examined to identify operating conditions, heat or mass transfer limitations.

The effect of incombustible matter, which occasionally or normally will remain in the waste (metallic scraps, buttons, glass debris, soil etc.) will also be investigated.

4.5.2.3. Off-Gas Treatment Parameters

This work will provide information for the selection and design of the off-gas treatment equipment. It will include measurement of the relevant off-gas properties, in particular the chemical composition, rates of evolution and the extent of particulate entrainment as well as a study of condensation and after burning off-gas.

Initially condensation and scrubbing will be applied to permit the measurement of the off-gas composition and evolution.

Condensation in two stages will yield heavy and light fractions for analysis, the first stage operating at about 100 °C, the second at near-ambient temperature.

The condensers will be followed by particulate filters and a caustic scrubber before exhaust.

After-burning will initially be studied in a separate rig using single components, synthetic mixtures and vaporised condensate samples. An engineered after-burner will be later tested in the programme.

4.5.2.4. Char Handling and Treatment

The following items will be studied :

- Characteristics of the char;
- Extraction of char from the reactor;
- Handling of low-density porous char in leaching equipment;
- Possible chemical reactions with leach solutions.

Consideration will be given to criticality and radiological problems.

4.5.2.5. General Plant Design and Operation

During the programme information relevant to plant design and operation will be collected and analysed to provide guidance in general areas, for example in selection of materials of construction, control systems and extraction of char.

This would include consideration of the performance of components under active routine and non routine conditions, their reliability and the ease of maintenance.

4.5.2.6. Equipment

In order to examine the chemical engineering features of the process a small batch pyrolysis unit will be operated during the first part of the programme. This unit has been designed and the equipment will be available for the start of the programme. After this first stage an improved design of pyrolysis reactor (Mark II) will be constructed and operated.

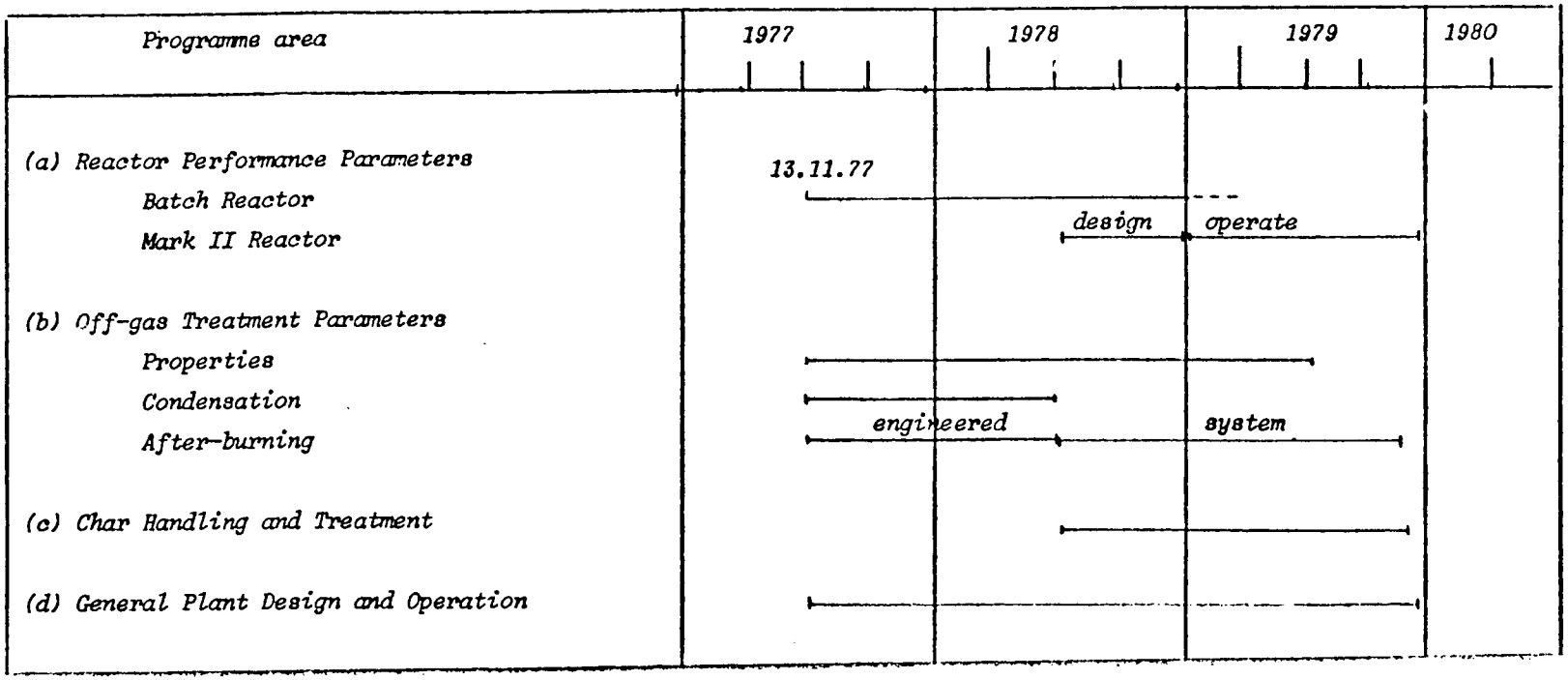
The type and design of the Mark II reactor will take account of the problems presented by active operation but will not be fully engineered to simulate an active unit.

This experimental unit will be operated during the last year of the programme.

4.5.3. State of the Programme

Background studies and basic experiments have been carried out at the UKAEA for some time and some of the results from these activities will be incorporated in this R & D project, which has officially started on 13 November 77.

Fig. 1 : Timescale of the Programme



4.6. Critical Factors in the Recovery of Plutonium (UKAEA, Harwell)

4.6.1. Objective

The aim of this programme is to provide the information needed to maximise the recovery of plutonium from combustible waste. Most effort will be devoted to the study of the effects of incineration and pyrolysis conditions and waste compositions on the leachability of plutonium from the ash.

4.6.2. Research Programme

4.6.2.1. Scope of the Programme

The programme consists of the following main actions :

1 - Construction and commissioning of an experimental furnace for producing "model"ashes;

2 - Preparation of model ashes from various synthetic wastes under different operating conditions;

3 - Investigations of relationships between extractibility of Pu and the variables : incineration/pyrolysis conditions, waste composition and form of Pu;

4 - Dissolution and characterization studies on pure Pu species - heat treated in absence of waste;

5 - Additional physical and chemical tests and treatments depending on the outcome of the previous series of experiments.

In addition to the samples prepared under this project, ash and residue samples from other EEC countries could also be included in the investigations, where meaningful.

4.6.2.2. Construction and Commissioning of an Experimental Furnace

A suitable furnace for producing "model" ashes from typical waste materials under fully pluto-

nium active and carefully controlled conditions will be constructed and commissioned.

4.6.2.3. Preparation of Model Ashes

Ash samples will be prepared from waste materials such as : PVC, Polyethylene, Neoprene and Hypalon rubber, cellulosic materials individually and as mixtures.

The wastes will be contaminated by : PuO_2, PuO_2/UO_2 mixed oxide, Pu-nitrate.

The furnace will be operated within the following ranges of conditions :

- Temperature : 400 - 1000 °C;
- Gas composition : Air - inert (e.g. N_2);
- Residence time : 10 min. - 5 days.

Detailed test schedules will be established after the commissioning of the new furnace.

Until this new furnace is in operation an existing small furnace will be used to prepare samples for dissolution tests.

4.6.2.4. Batch Dissolution Tests and Characterization Studies

Each sample of ash will be subjected to a standard batch dissolution test to determine the extent to which plutonium can be extracted. In cases where the standard dissolution test appears to give less recovery than could be expected with other reagents or test conditions, the possibilities for optimum extraction of Pu will be investigated. In addition to these dissolution tests, selected samples of ashes will be characterized by other measurements, e.g. chemical analysis, ceramography, X-ray diffraction, spectrographic analysis.

In this way information will be obtained relating the extractability of plutonium to the incineration/pyrolysis conditions, the nature of the waste and the form of the plutonium contamination.

4.6.2.5. Tests with Pure Plutonium Species

Dissolution and characterization studies will also

be performed on pure plutonium species, which have been subjected to conditions related to those of the incinerations but in absence of waste material. The species (PuO_2, PuO_2/UO_2, Pu nitrate) and the conditions (temperature, gas composition and residence time) will essentially be the same as those for the model ashes.

4.6.2.6. Complementary Studies

In the light of the results from the above work, consideration will be given to additional physical and chemical treatments that may enhance the extraction of the plutonium from the ashes.

4.6.2.7. Tests with non UKAEA Ash Samples

Where appropriate dissolution and characterization tests could be applied to samples produced in other plutonium active incinerators. It would be desirable to have well defined conditions of incineration or pyrolysis and necessary to have all relevant information on these conditions and the nature of the wastes made available for this programme. The cost and responsibility for the transport of the materials would be incumbent on the organisation producing the ash.

4.6.3. State of the Programme

Although considerable preparatory work has been carried out at Harwell in 1977, the EC research contract will only cover activities from 13 November 1977 on words. At the end of 1977, the contract negotiations had been completed and the administrative procedure is due to begin early in 1978.

TIMETABLE

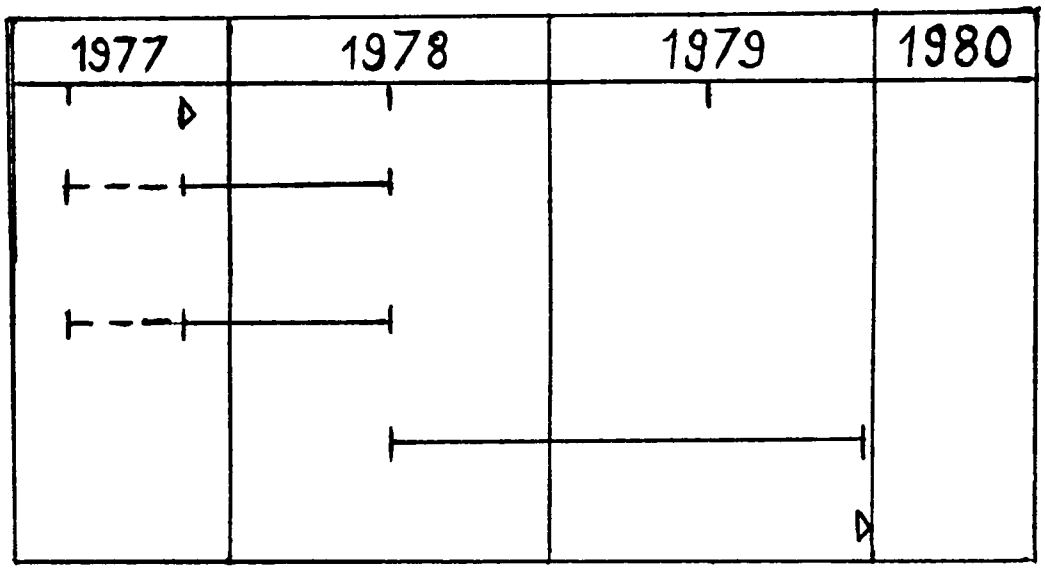
Start of contract

Construction and commissioning of the new furnace

Exploratory studies : examination, dissolution and characterization of ash samples prepared in the small, experimental furnace

Main test series : systematic study of ashes from the new furnace

End of contract



4.7. Radiation Assisted Incineration Study (Gravatom-UK)

4.7.1. Objective

The aim of the R & D contract with Gravatom is to develop a technique to incinerate plutonium contaminated waste at low temperature and under the assistance of ionising irradiation so as to reduce combustion hazards under conditions which favour subsequent plutonium extraction.

4.7.2. Technique

To provide maximum plutonium retrievability, the degree of oxidation during a combustion process should be kept low. This could be achieved in a practicable way by a low temperature oxidation (at 200 - 300 °C) assisted by ionising radiation. The latter lowers the initiation temperature for the chemical reaction. Once there is a sufficient density of reacting molecules an exothermal reaction becomes self sustaining due to the ready availability of new reactive molecules.

4.7.3. Scope of the Programme

The programme is divided into two stages :

Stage I : A feasibility study of the process backed up with low level irradiation tests. The study started in November 1977 and is scheduled to be completed at the end of June 1978. It will then be evaluated by the EC-Working Party for "Incineration of plutonium contaminated waste".

Stage II : The construction and 6 months of operation of a laboratory scale experimental plant are scheduled for 1979. The detailed working programme of stage II will be defined after examination of the feasibility analysis. A timetable of the working programme is attached.

4.7.3.1. Feasibility Study

The feasibility analysis consists of :

- The inventory of potential waste arisings and

the waste compositions;

- The study of the process on aspects like : ignition temperature and oxygen level, rates of reaction and exposure time, catalysis, ionisation heating and radiation level and the intended properties of the pyrolysis and radiolysis products;
- The identification of specific aims for research and the design of a laboratory scale experimental plant with particular emphasis on criticality, safety and reliability aspects.

4.7.3.2. Construction of the Experimental Plant

The plant will be designed for a capacity of approximately 100 g/h. The combustion chamber shall be installed in a high flux irradiation device.

4.7.3.3. Experimental Work

The plant will be operated to investigate the following parameters :

- The initiation point of oxidation for individual materials and various levels of oxygen in nitrogen;
- The degree of depression of the threshold temperature under high ionisation fluxes;
- The heating effect of high ionisation fluxes;
- The effectiveness of catalysts if any;
- The composition of the off-gases.

The objective of these experiments is to find the best combustion conditions with respect to the reduction and chemical stability of the residues and with respect to plutonium extraction.

The following of the residues will be measured : inflammability, volume- (weight-) reduction, density, chemical composition and plutonium extraction rate (by simulation).

4.7.3. . Evaluation of the Process

The experimental results will be evaluated and the application of the process will be discussed, further R & D work required and relevant modifications of the experimental plant to allow handling of plutonium contaminated waste will be defined.

TABLE 1

Time schedule for the research programme "Low temperature incineration of plutonium contaminated waste assisted by gamma-irradiation"

Actions	1977 IV	1978 I	1978 II	1978 III	1978 IV	1979 I	1979 II	1979 III	1979 IV
1 Stage I : Feasibility study	1.Nov.			Special Report					
2 Stage II :									
2.1 Construction of experimental plant				Expert Meeting			1. Interim Report		
2.2 Experimental operation of plant and							2. Interim Report		
2.3 Examination of residues									
2.4 Evaluation of results									Final Report

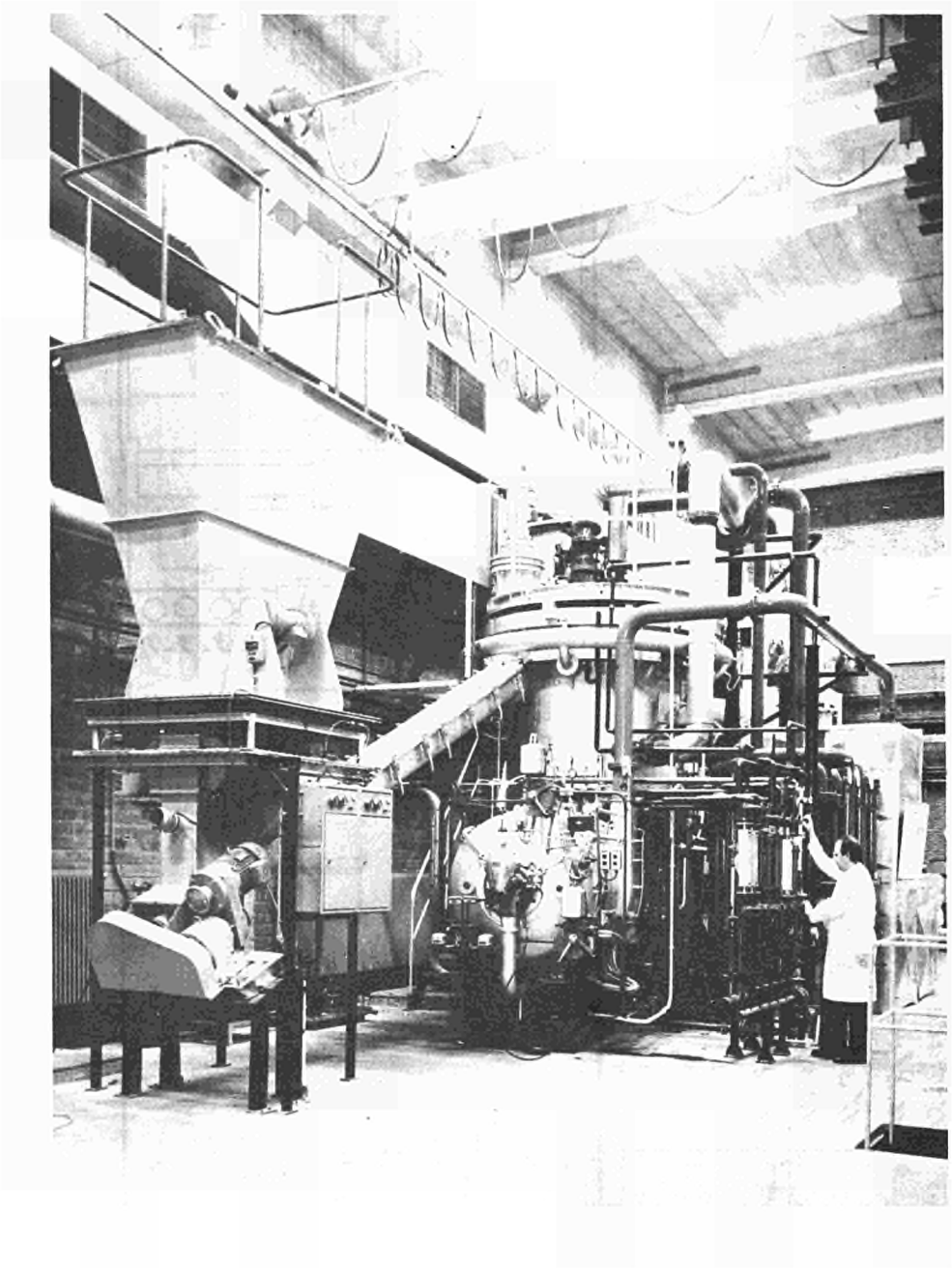


FIG. 4.2.1 : View of the FLK 60 plant

1. container liftsystem.
2. waste transfert room.
3. sorting area.
4. balance.
5. crusher.
6. mixing silos.
7. F.L.K. 60.
8. heat exchanger n°1.
9. panelbedfilter.
10. heat exchanger n°2.
11. scrubber unit.
12. final abs. filtration.
13. ash scraper.
14. ash incorporation.
15. material transfert room.
16. sludge drying.
17. sludge tichner.
18 sludge tanks.
19. crane manipulator.
20. frogmen access.
21. controle room.
22. separation wall. α zon. cont. room.
23. separation wall. α zon. entry.

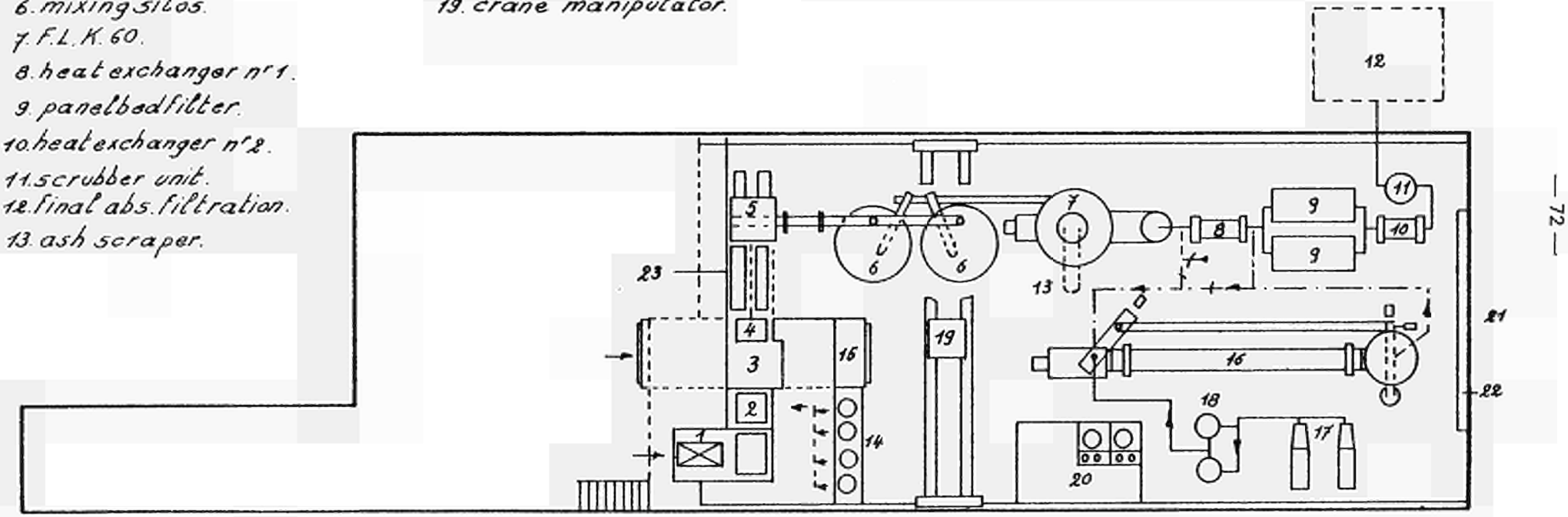


FIG. 4.2.2 : Inplantation of the different components in the F.L.K. Plant Hall.

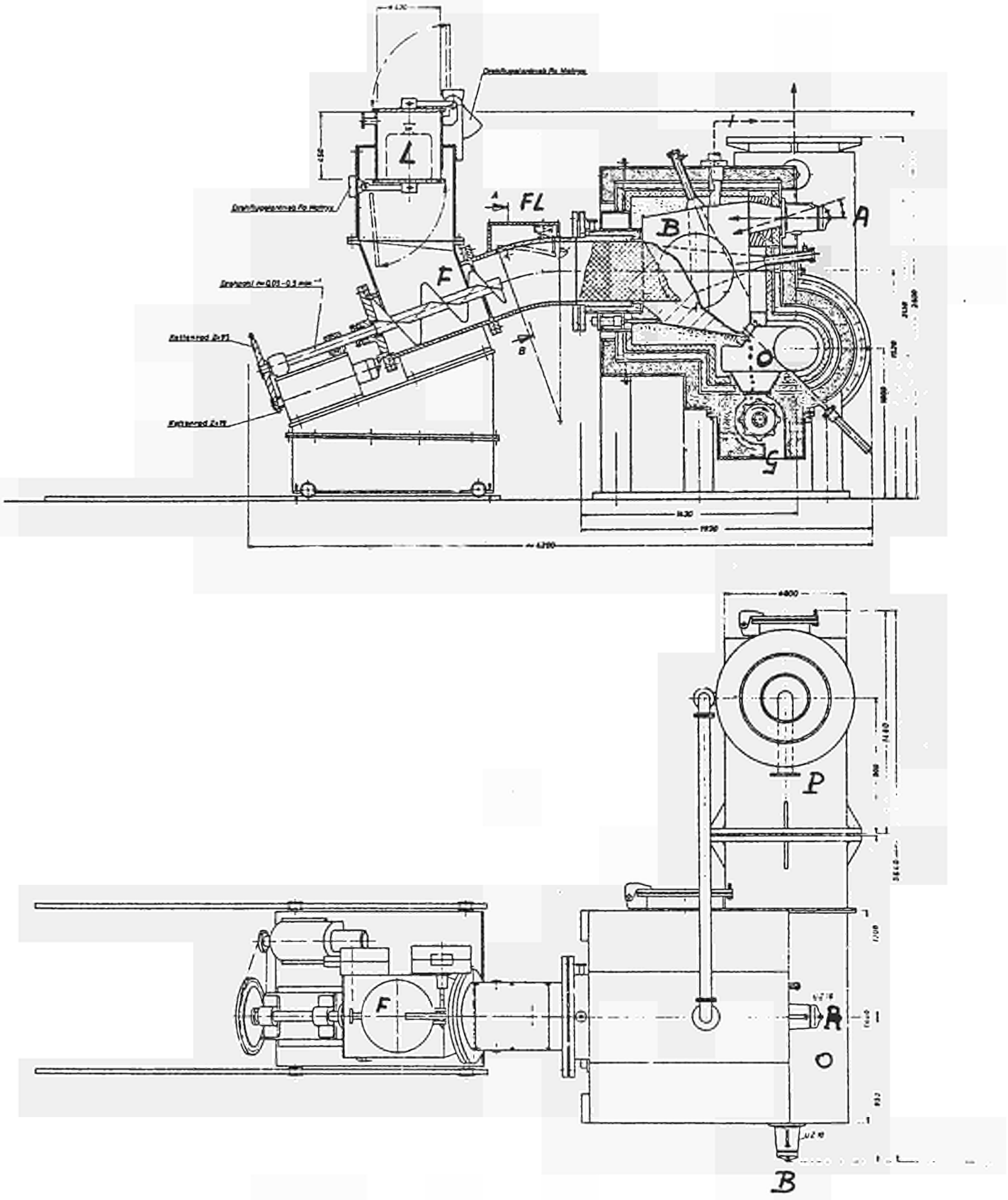


FIG. 4.2.3: Incinerator for Pu waste (10 kg/h)

RU16

R

10^{-2} 10^{-3} 10^{-4} 10^{-5} 10^{-6} 10^{-7} 10^{-8} 10^{-9} 10^{-10}

64.00 96.00 128.00 160.00 320.00 352.00 384.00

T (DAYS)

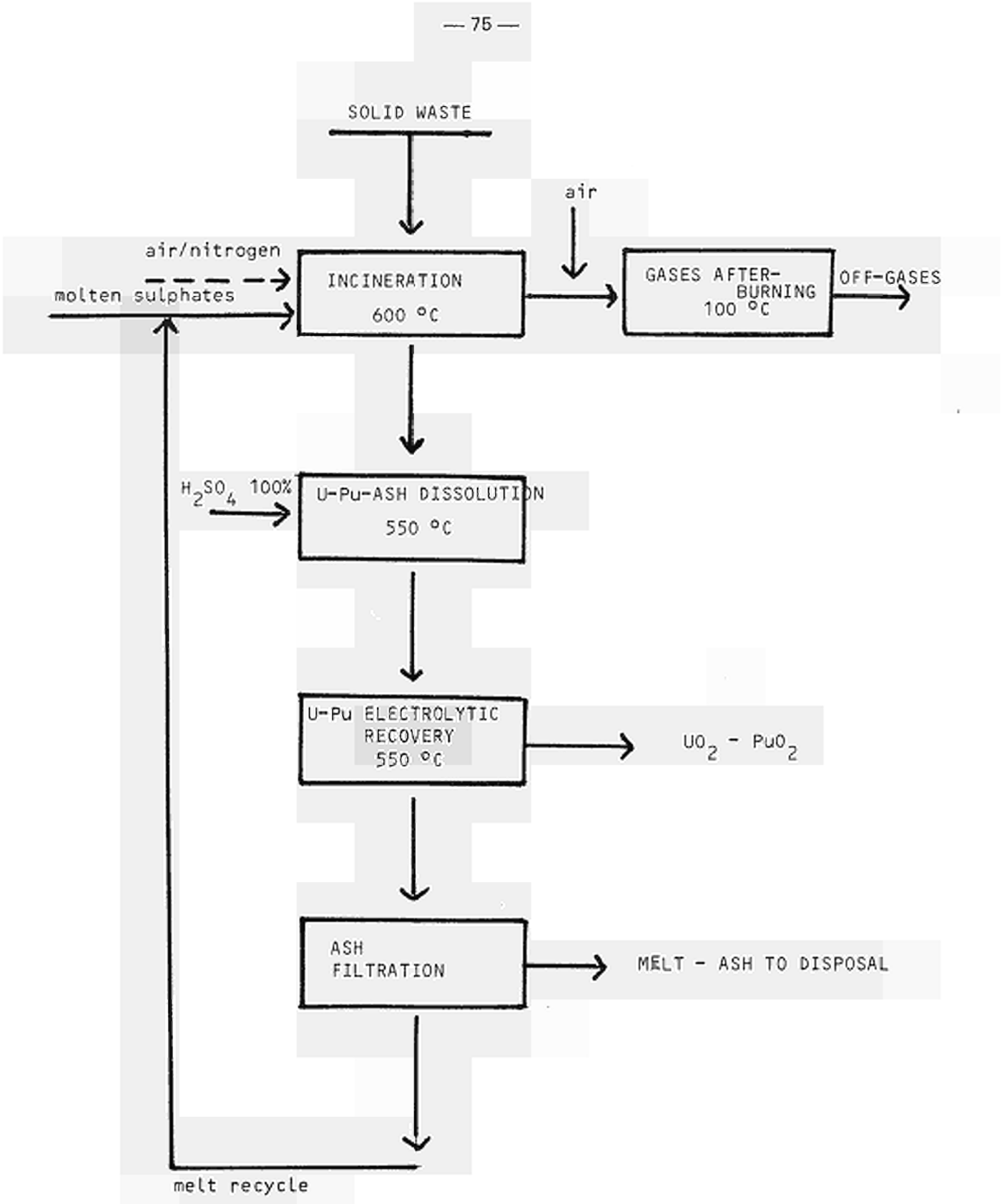


Fig. 4.3.1 - Flow-sheet of the molten salt incineration process

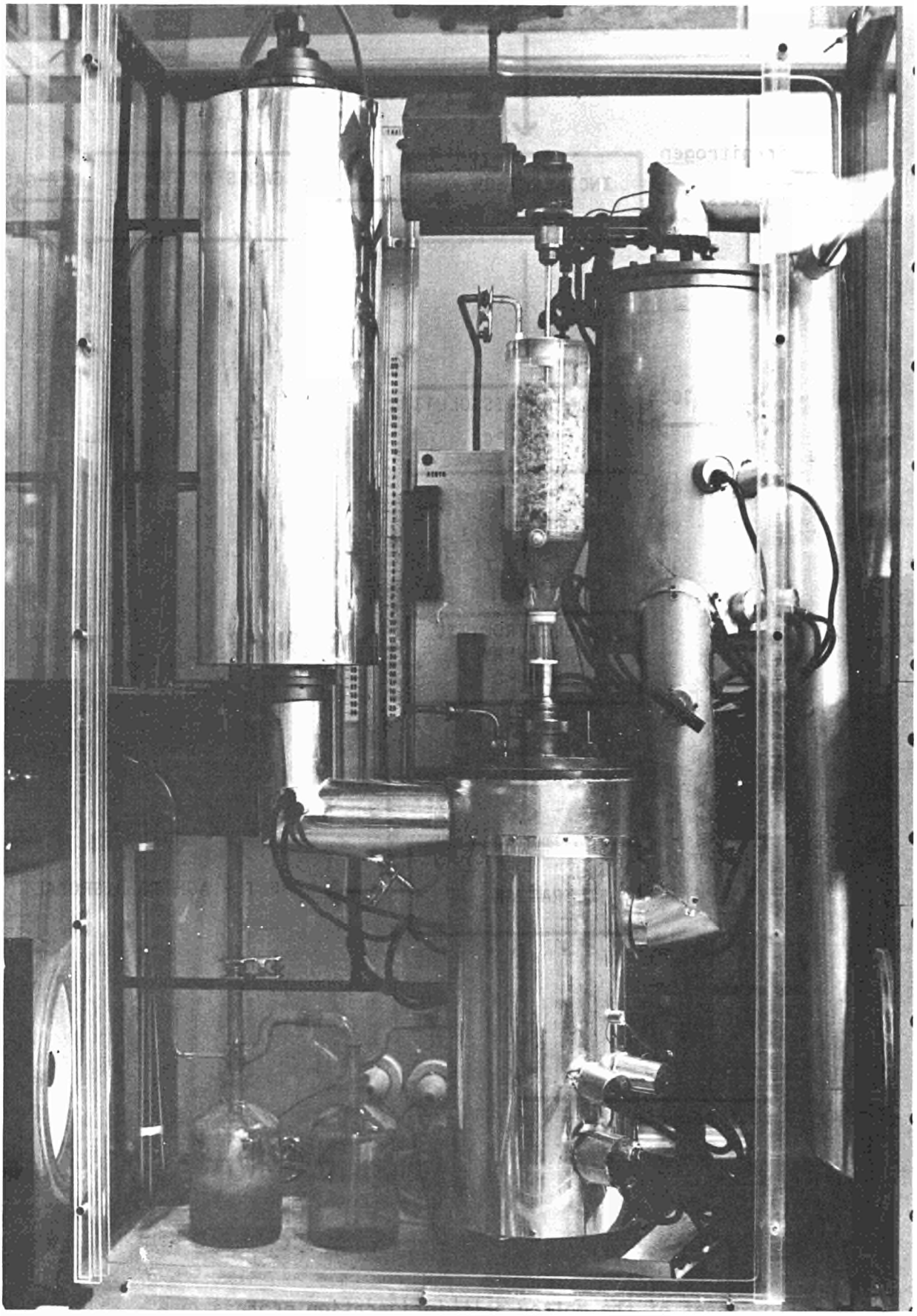


Fig. 4.3.2 Bench-scale Test Rig

Fig. 4.4.1 Project Wet-Combustion

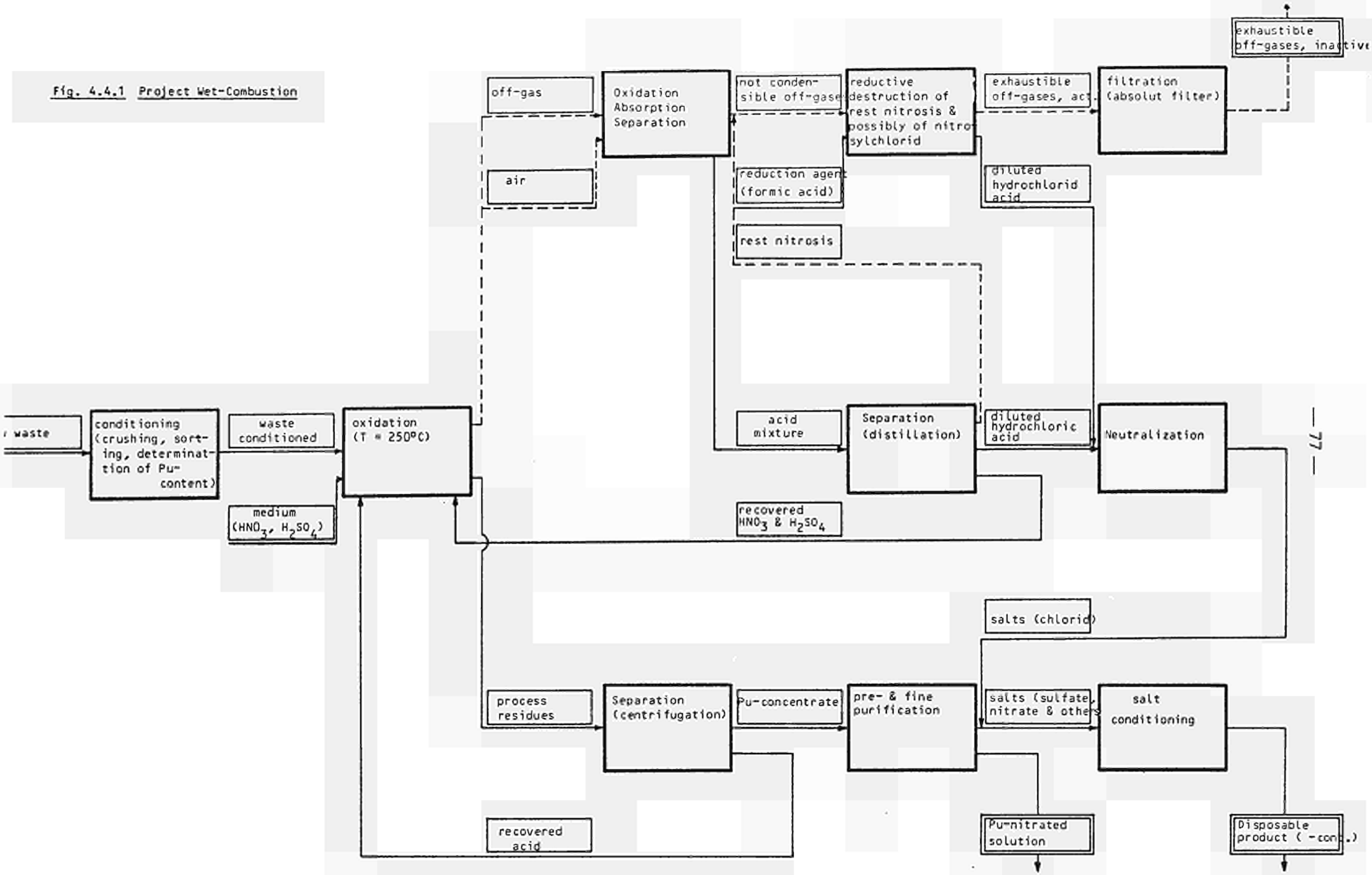


Fig. 4.4.2 Mass flow sheet of inactive laboratory plant ELONA

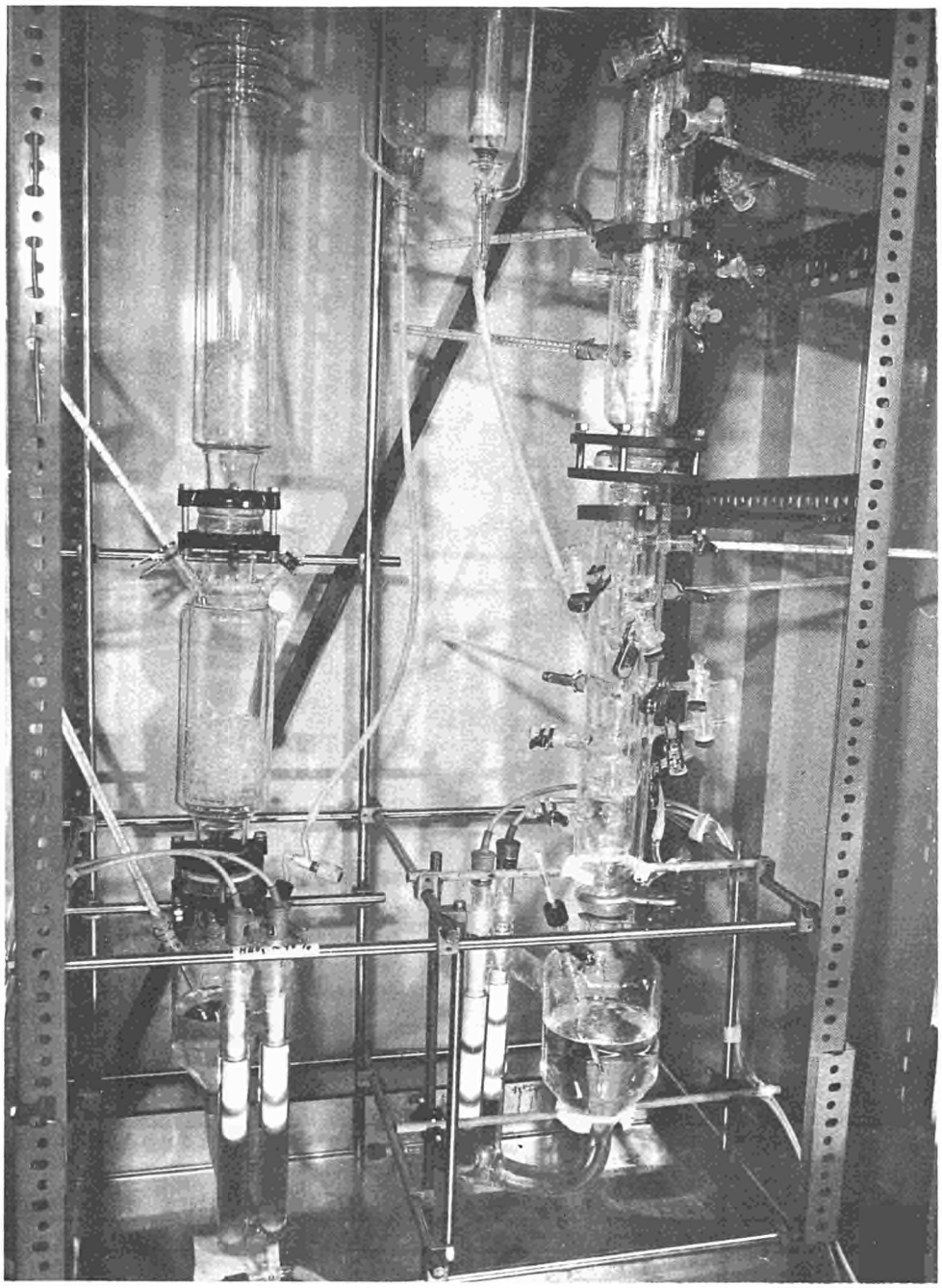


Fig. 4.4.3 Rectification column with filling (left) and interim steps (right) for determination of the layout of the acid separations unit

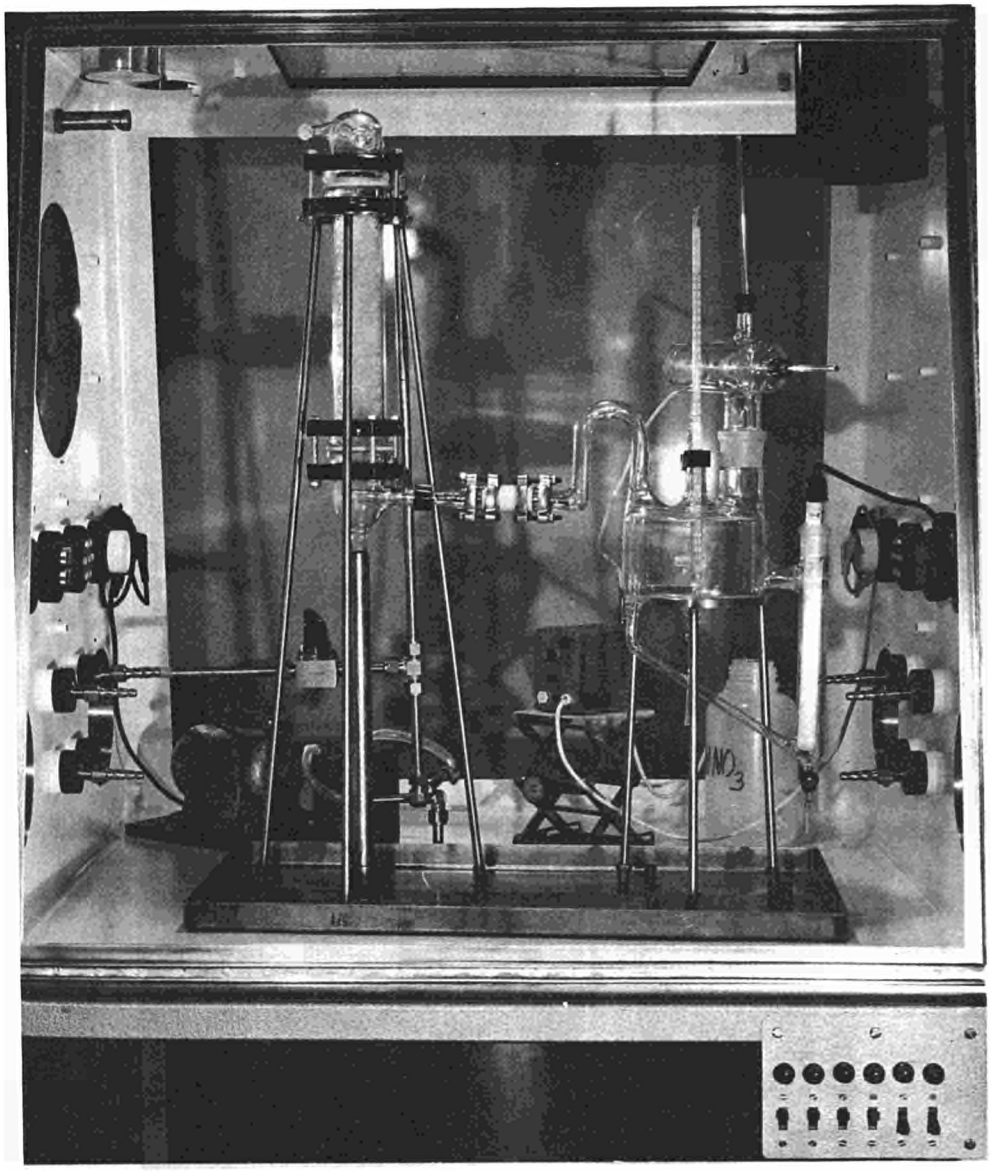


Fig. 4.4.4 Glovebox with equipment for the wet-combustion (small scale), combustion chamber (right), off-gas wash column (left)

5. TESTING AND EVALUATION OF SOLIDIFICATION SUBSTANCES FOR HLW

(Joint action UKAEA-HMI-CEA)

5.1. Introduction and Objectives

The most active work of reprocessing is the High Level liquid waste resulting from the dissolving of spent fuel in nitric acid. Transport long term storage and final disposal of this work must be preceded by their incorporation into a solid insoluble material.

Glasses are presently the preferred materials and the development of the solidification processes has advanced to a stage where one active prototype plant and a number of pilot plants are being constructed in Europe.

The coordinated action of the United Kingdom Atomic Energy, the Hahn-Meitner Institut and the Commissariat à l'Energie Atomique intend to establish a joint basis for the evaluation of solidification substances.

The programme concentrates on series of experiments jointly testing five (probably six from 1978 onwards) different solidification substances from the three participating countries, performed under strictly identical conditions.

The primary aims of the programme are :

- Determination of the leach resistance under various conditions
- Investigation of the effects of devitrification
- Investigation of the effects of alpha radiation.

5.2. Devitrification Effects on Physical and Chemical properties of HLW solidification Substances (Hahn-Meitner-Institut Berlin - D)

5.2.1. Objectives

The Hahn-Meitner-Institut (HMI) will investigate the effects of heat treatment on the physical and chemical properties of HWL solidification substances (glass products). In particular crystallization of glass products will be investigated in detail. The preparation of the four reference glasses and the German glass ceramic as well as the heat treatment are performed by UKAEA Harwell. The five substances have been annealed at 500, 600, 700 and 800°C for the durations of 2, 24, 240 and 2400 hours. Approximately half of the samples have been received at HMI up to now.

Investigations were started with samples annealed at 800°C for 2,24 and 240 hours.

5.2.2. Completed work - Results

5.2.2.1. Leach experiments according to the grain titration method DIN 12111

The water resistance of the products under investigation has been measured using the grain titration method.

Preliminary results are given in table 5.2.1. The second column of the table gives values for the "as prepared" (stress released) glasses. With the exception of the French glass 58.30.20, no significant changes in the alkali release were observed in comparison with the as prepared samples. For the French glass the alkali release increases by half the value for annealings at 800°C and 24 hours, and by a factor of ~ 5 for annealings at 800°C and 240 hours. It should be noted that there is generally a direct relation between alkali content and release.

Table 5.2.1. : Results of the determination of water resistance according to DIN 12 111.

Glass	Hydrolytic resistance after heat treatment Alkali release [µeq/g]			
	1 h 500°C	2 h 800°C	24 h 800°C	240 h 800°C
Glass F 58.30.20	20.4 20.6	25.4 26.9	37.9 37.2	127.5 129.4
Glass ceramic B1-3	1.5 1.3	3.1 3.4	2.9 3.1	2.7 2.5
Glass VG 98/3	37.0 36.6	38.3 35.5	41.5 41.6	37.8 37.8
Glass UK 209	2.3 2.3	1.9 1.9	1.9 1.8	1.9 1.7
Glass UK 189	8.7 8.7	6.9 6.9	6.5 6.5	6.0 5.9

5.2.2.2. Differential thermoanalysis (DTA) and X-ray diffraction experiments

DTA traces were performed with the four glasses and the parent glass of the German glass ceramic. The influence of the heating rate on peak position and size was measured by using linear velocities of 1,10 and 100°C/min up to 1000°C. The results are presented in Fig. 5.2.1. The glass 58.30.20 shows some more or less weak exothermal peaks near 740°C. A subsequent X-ray analysis of the DTA sample showed significant traces, i.e. the glass was partially crystallized upon heating. The parent glass B1-3 shows, as expected, the strongest DTA peaks of all samples as well as strong X-ray traces. The glasses VG 98/3, UK 209 and UK 189, show a less pronounced tendency to crystallize. Only VG 98/3 shows some weak X-ray traces in the case of the specimen heated with 1°C/min. However, crystal phases were formed in all cases.

5.2.2.3 The energy dispersive microprobe system (DEX)

Fig 5.2.2 shows SEM-pictures of polished and with gold vapourized surfaces of the glass ceramic B1-3. The various crystal phases are well recognizable. Fig 5.2.3 shows the spectra of the as prepared and Fig. 5.2.4. of the remaining glass phase of the ceramic B1-3. The comparision of the peak intensities of both spectra shows a pronounced decrease of elements Cs, Ba, Ti and the rare earths, which are reappearing in a concentrated form in the newly formed crystal phases. This implies, that the composition of the remaining glass phase after crystallization is distinctly different from the initial product which together with the herein finally dispersed crystals entails a deviation of properties of the entire product.

5.2.2.4 Wavelength dispersive microprobe system (WDX)

A higher resolution of X-ray energies as compared to the EDW can be obtained with the wavelength dispersive system Fig. 5.2.5. The device is equipped with two spectrometers and a phase integrator. Fig. 5.2.6. gives an image of the resolution of the WDX-system for the example shown in Fig. 5.2.3. in the energy region of 4 to 5 keV.

5.2.2.5. Scanning electron microscopy (SEM) and electron probe analysis (EPA)

SEP-pictures and first element distribution pictures have been produced for all but the 2400 hrs products annealed at 800°C. As an example SEM-EPA-pictures are shown for the French glass 58.30.20. The SEM-picture of the 240 h/800°C specimen in Fig. 5.2.7. shows dis-

tinctive crystallization features. The white circular phase in the picture above-left and the large bright phase at the right hand edge of the picture are the nobel metal phases of Pd i.e. Ru, which were already present in an unsolved form in the initial glass. Apart from that, there are at least two further phases present. According to the present state of evaluation it only can be said that molybdenum is concentrated in the fine scattered white phase (see Fig. 5.2.7.b), whereas silica is concentrated in the dark phase (see Fig. 5.2.7.c). The dark contrast of this phase implies that it is a phase with a low ordinal number. This leads to the presumption that it is an easily soluble alkali silicate, which would explain as well the increase of the alkali release as indicated in table 5.2.1.

5.2.2. Interim conclusions

It can be concluded, from the preliminary evaluation, that all glass products showed microscopic inhomogeneities in their as prepared state and that none of the products dissolved the noble metals entirely. On all four glasses annealed at 800°C multiphase-crystallization was observed. The formation of crystal phases seems to depend on the annealing temperature and time. The distribution of individual elements in the crystal phases depends on the glass composition.

5.2.3 Planned activities

All but the 800°C series of annealed samples are to be prepared, leached and crystallographically investigated. The evaluation of results is performed in parallel.

5.2.4. References

EUR 5749 - Annual progress report 1976 of the radioactive waste management programme (indirect action).

5.3. Analysis of the Effects of Leaching and Radiation (UKAEA-HARWELL-GB)

5.3.1. Objectives

The UKAEA contribution to the joint programme is aimed at the comparative testing, under the same conditions, of the five reference substances (for solidification of HLW) with respect to temperature and irradiation dependent damaging effects. The programme comprises :

a) The manufacture of non-active samples of the five substances.

b) The annealing of parts of these samples, at various temperatures and for various times to investigate any tendency towards crystallization.

c) The leach testing of the samples ; annealed samples at 100°C only and the as-cast state as well at various lower temperatures.

d) The testing of the irradiation stability of the glasses by incorporating 238Pu 0_2 so that a dose equivalent to several hundred years storage is accumulated in 2 years.

5.3.2. Completed work

5.3.2.1. Production of inactive samples

3 kg of each of the five reference substances were manufactured according to the formulae supplied by the collaborating laboratories. The compositions used are summarized in table 5.3.1. The formation temperatures of the five substances are as follows :

UK glass 209	1000°C
UK glass 189	950°C
French glass SON.58.30.20.42	1150°C
German glass VG 98/3	1200°C
German glass ceramic B 1/3	1200°C

Some segregation of "yellow phase", an alkali molybdate-chromate, was observed after melting the glass ceramic. The Hahn-Meitner-Institut (HMI) pointed out that the constituants of glass-former and waste, instead of mixing them directly together, the glass-formers should have been premelted to form a frit and the waste added subsequently. A second batch, made in this way was homogeneous.

5.3.2.2. Heat treatment of samples

To investigate the tendency of the glass products to devitrify, samples are to be annealed at 500, 600, 700 and 800°C for periods of 2, 24, 240 and 2400 hours. Most of the heat treatment has been completed and some of each batch has been sent to HMI for crystallographic examination.

5.3.2.3. Preparation of radiation stability tests

It is generally agreed that the most significant radiation damages on waste glasses are caused by alpha-radiation. It was thought most appropriate to investigate the radiation stability of glasses by doping them with an excess of an α-emitting isotope (^{238}Pu) so that the samples would receive in two years as many α-decays per gram ($\sim$ 8,8 x 10^{17}) as the real vitrified waste will in several centuries.

Accordingly four samples of each of the five substances have been spiked with 2,5 wt. % ^{238}Pu, replacing the rare earth elements in the fission product spectrum to minimise chemical effects in the glass. Autoradiagraphs have been made of the samples and the initial leach rate and density has been measured. After two years storage, repeat measurements will show probable changes. The stored energy (Wigner energy) and the amount of helium release from the samples will then also be measured

5.3.2.4. Results on leach testing

All leach tests are carried out by the Soxhlet method. The leach rate is determined by weight loss. The surface layer, which is deficient in alkalis, boron, etc., was separated off before the final weight was measured. Five leach tests at 100°C were carried out on seperate specimens of each of the inactive and as-cast substance. The average results were :

Glass Product	Leach Rate $[g\ cm^{-2}\ day^{-1}]$
UK 209	$2,6 \pm 0,9 \times 10^{-4}$
UK 189	$1,3 \pm 0,2 \times 10^{-3}$
SON 58.30.20.42	$3,1 \pm 0,3 \times 10^{-3}$
VG 98/3	$1,9 \pm 0,3 \times 10^{-3}$
B 1/3	$6,6 \pm 0,6 \times 10^{-4}$

Leach tests at temperatures below 100°C(50, 60, 70, 80, 90°C) are in progress.

The first annealed samples, series 800°C for 2,24 and 240 hours, have been leach-tested at 100°C, (the results of which are given in Fig. 5.3.1.).

5.3.2.5. Interim conclusions

(The increase in leach rate of the French glass after 10 days at 800°C (Fig. 5.3.1.) is not of great significance for its behaviour during preparation or in storage, since it is unlikely that it would be held for so long at such a high temperature).

5.3.3. Planned activities

The manufacture of all samples has been completed. Heat treatments at 500°C for 24 h and at 600 and 700°C for 2 400 h well as leach tests for

all but the 800°C-series have yet to be carried out. At the end of 1979 the radiation stability of the Pu-doped samples will be examined.

Table 5.3.1. : Compositions of the 5 glasses.

	UK 209	UK 189	French SON 58.30.20.U2	German Glass VG 98/3	German Celsian Glass Ceramic B1/3
GLASS FORMERS					
SiO_2	50.88	41.51	43.6	41.84	28.0
B_2O_3	11.12	21.87	19.0	10.48	6.4
Na_2O	8.30	7.68	9.4	20.24	1.6
Li_2O	3.99	3.69			2.4
TiO_2				3.52	4.0
CaO				2.32	4.0
MgO				0.4	1.2
Al_2O_3				1.2	12.8
ZrO_2					0.8
As_2O_3					0.4
BaO					14.8
ZnO					3.6
WASTE					
FPO_x	9.75	9.58	22.71	15.64	15.11
U_3O_8 } ($4C_2$)	0.06	0.06	3.6	1.21	0.47
AcO_2 }					
Fe_2O_3	2.73	2.68	0.6	0.70	1.51
Cr_2O_3	0.56	0.55	0.2	0.24	0.43
NiO	0.36	0.36	0.1	0.21	0.24
Na_2O				2.01	2.23
Al_2O_3	5.11	5.03	0.1		
MgO	6.34	6.23			
ZnO	0.44	0.44			
P_2O_5 (PO_4)	0.24	0.23	0.6		
SO_4	0.10	0.09			

5.4. Leach Tests with Fission Product bearing Glass Blocks (CEA Marcoule - F)

5.4.1. Objective

CEA will measure the leach rate of five reference samples with volumes of approximately 0,8 litre each, containing up to 20 % of LWR fission product oxydes (activity 100 to 1000 Ci/3 γ). The following leach rates will be measured by radioanalysis daily and at room temperature during about 40 days :

- Total leach rate of beta- and gamma- emitters,
- Specific leach rate of beta- and gamma- emitters of the isotopes :

  ^{137}Cs, ^{90}Sr, ^{144}Ce and ^{106}Ru,

- Total leach rate of alpha-emitters, if measurable.

The leaching agent is tap water (volume : 700 ml) with a resistivity of 2500 Ω/cm at 20°C and a pH-value of 7,5.

5.4.2. Planned activities

The programme starts at the beginning of 1978. Viscosity tests are being carried out during January on the five reference glasses. The manufacture of the block samples is anticipated to start in February.

5.5. Complementary Actions

The following complementary actions have been introduced during 1977 into the programme :

5.5.1. Effects of different leaching media and of the ionisation of the leachant (UKAEA, Harwell-GB)

5.5.1.1. Different leaching media

It has been found that the leach rates in water containing various naturally occurring impurities can vary markedly from the values found in distilled water. The effect is usually to reduce the leach rate but some glasses leach more rapidly in acidic (low pH) water. According to this the glasses are being leach tested in the following media at temperatures of 60 and 90°C :

a) Sea water/salt water,

b) Water in equilibrium with clay and granite,

c) Waters with pH values of 3, 5, 7 and 9.

For the leach tests in clay water, clay samples from the potential disposal site of Belgium (Mol) have been supplied.

5.5.1.2. Ionisation of the leachant

Another factor which may affect the leach rate is the ionisation of the leachant due to the radioactivity of the glass. This can be investigated either by using a gamma-irradiation facility or by irradiating the leachant and leach specimen with X-rays. The latter method was preferred since it is then easier to control the pH-level and the temperature of the leachant. These tests will be carried out at 90°C, comparing results under the same conditions but without X-rays and probably also with different X-ray fluxes.

5.5.2. Leach tests with fission product bearing glass blocks at various temperatures (CEA Marcoule-F)

The leach tests which CEA is performing with the five active block samples at room temperature will be repeated at 50 and 85°C in order to derive a more comprehensive set of data for the comparison with the inactive leach tests performed in parallel at UKAEA Harwell (see chapter 5.3.2.4.).

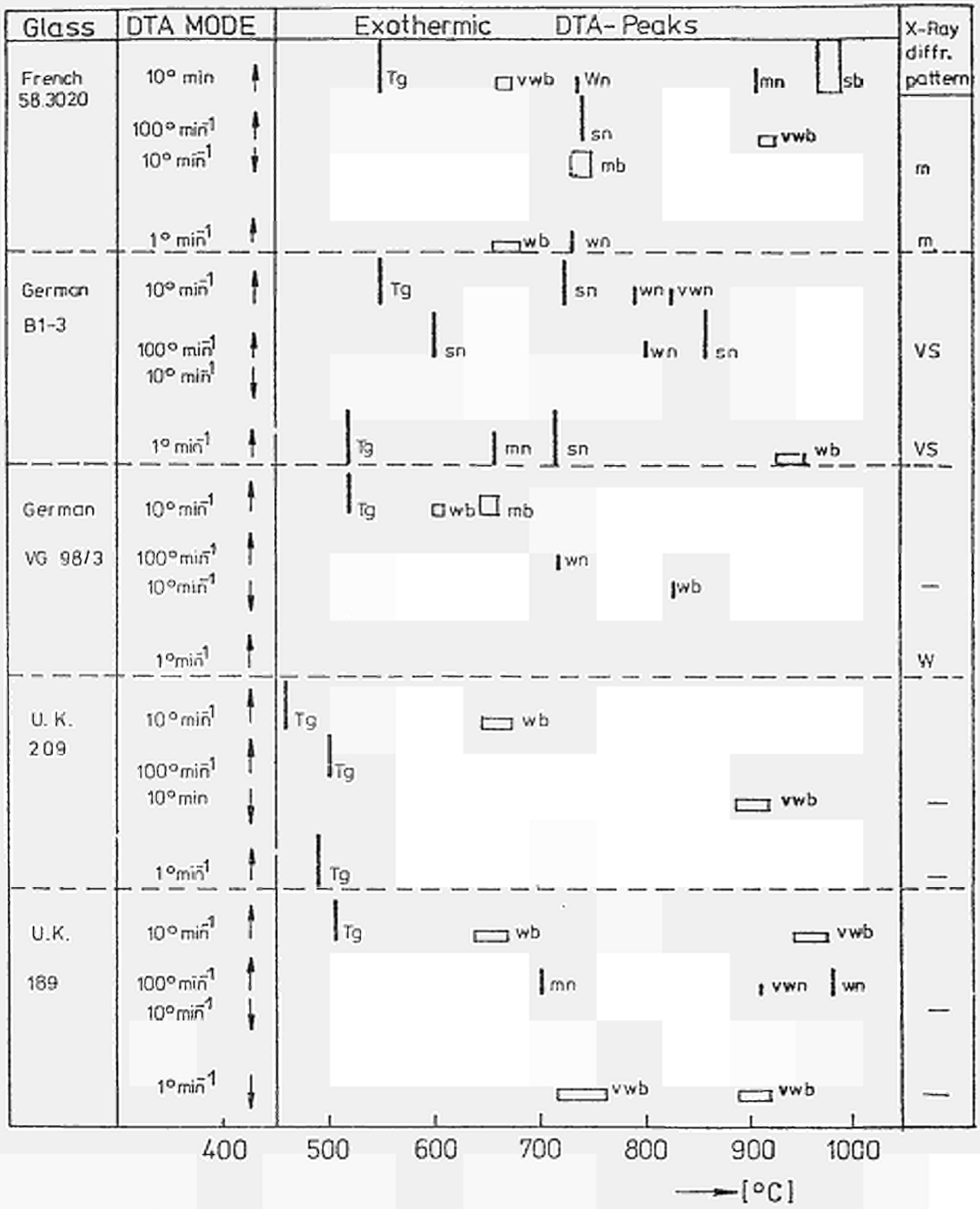


Fig. 5.2.1 Schematic representation of DTA experimental results

Tg = temperature of transformation

Relative intensity and shape of the peaks:

vw	= very weak	b	= broad
w	= weak	n	= narrow
m	= medium		
s	= strong		

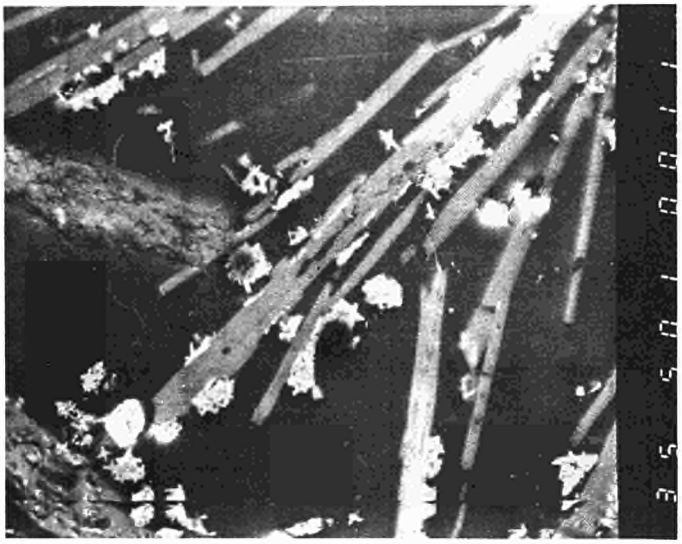


Fig. 5.2.2a

Magn. 500 x
Spot in Phase 1

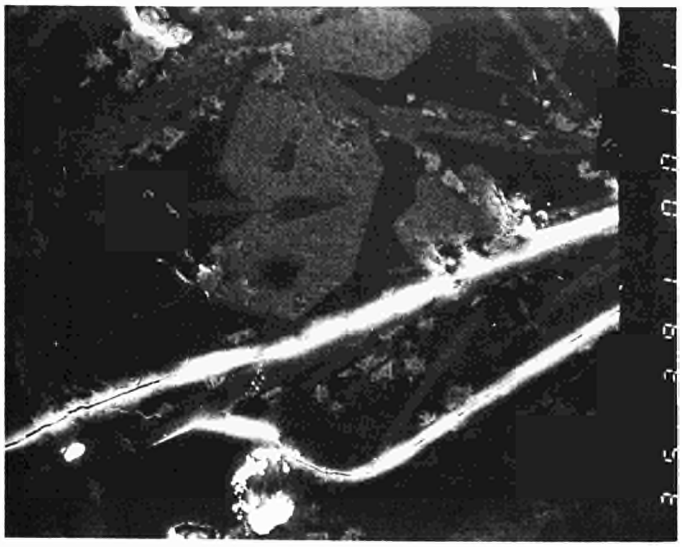


Fig. 5.2.2b

Magn. 390 x
Spot in Phase 2

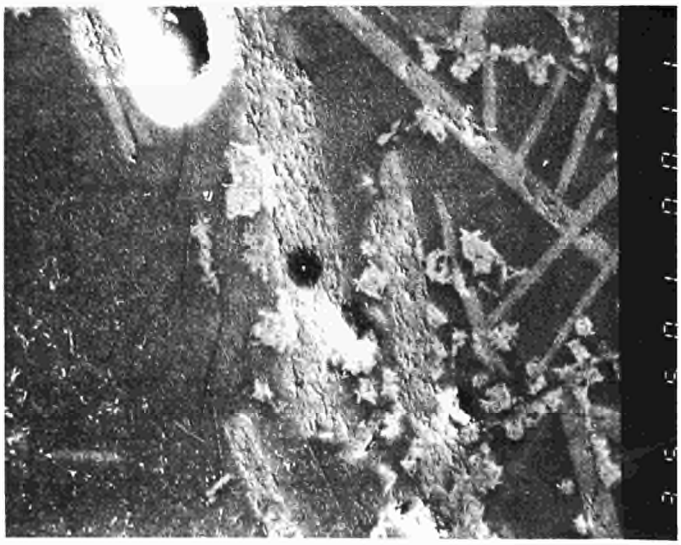


Fig. 5.2.2c

Magn. 500 x
Spot in Phase 3

Fig. 5.2.2 SEM-photographs of a polished surface of the glass ceramic Dl- 3

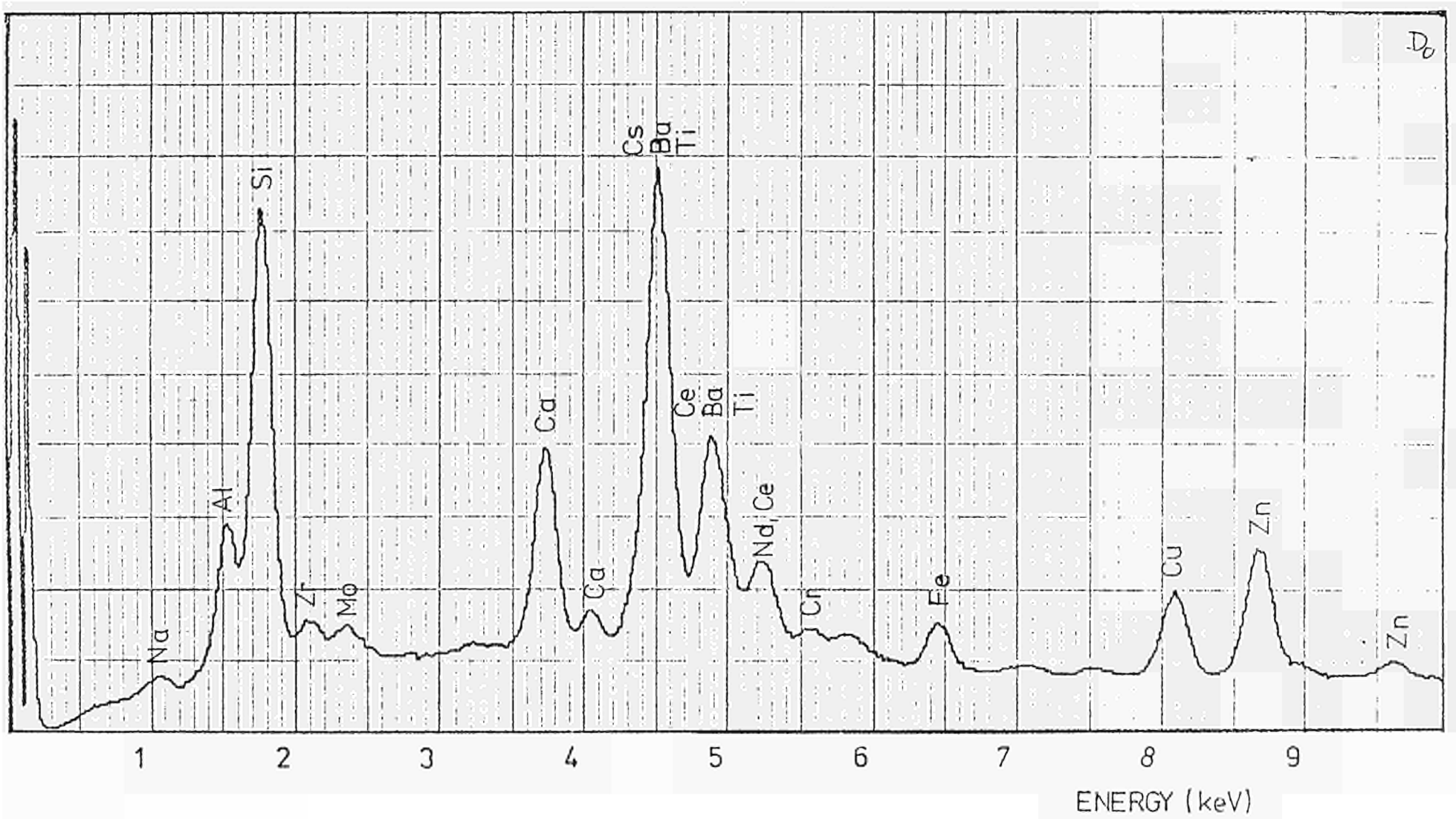


Fig. 5.2.3 Energy dispersive x-ray fluoreszent spectra of the initial glass of the glass ceramic B 1-3

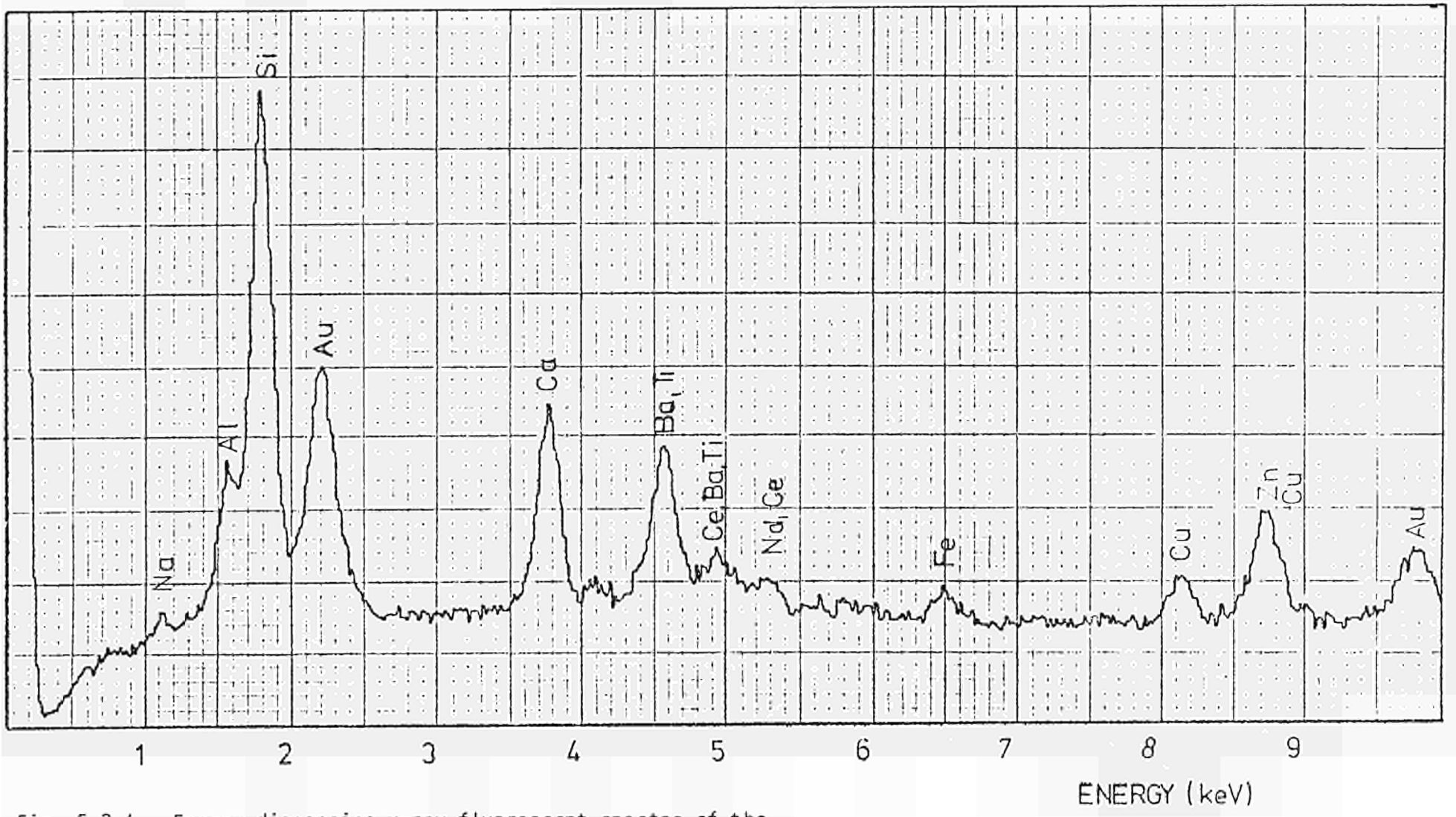


Fig. 5.2.4 Energy dispersice x-ray fluorescent spectra of the remaining glass phase of the glass ceramic B1-3

Fig. 5.2.5 Scanning electron microscope JSM U3
with wavelength dispersive double X-ray spectrometer,
image analyser and video tape recorder.

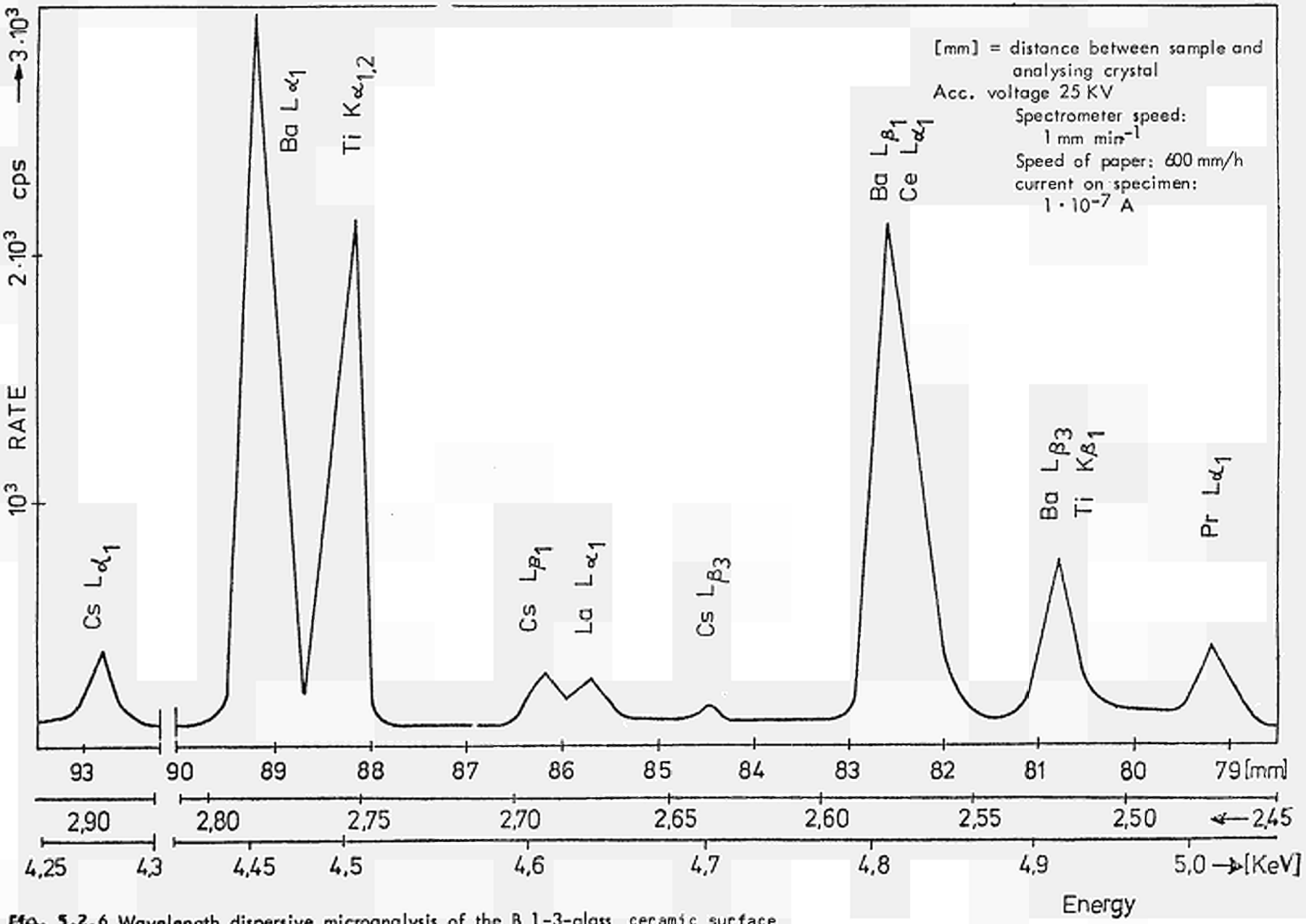


Fig. 5.2.6 Wavelength dispersive microanalysis of the B 1-3-glass ceramic surface

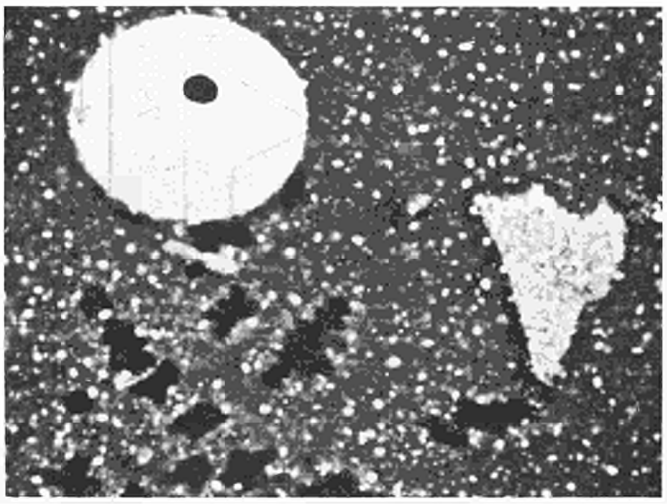


Fig. 5.2.7a
SEM-picture of the heat-treated glass F 5830.20
Annealing temp. 800 °C
Annealing time 240 h
Magn. 1000 x

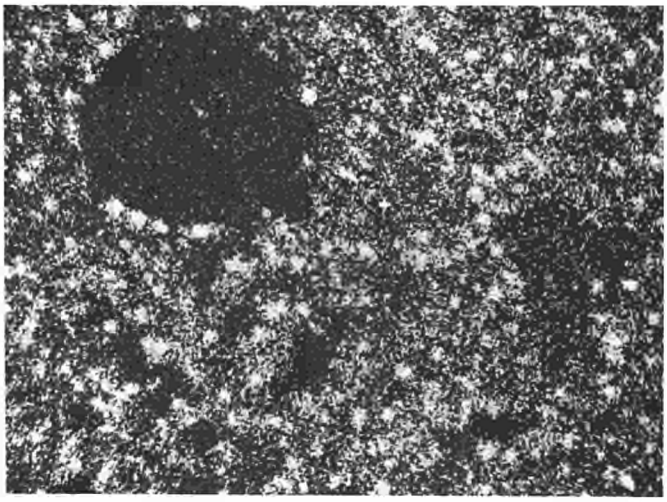


Fig. 5.2.7b
EPA-Element distribution of Mo in Fig. 5.2.8a

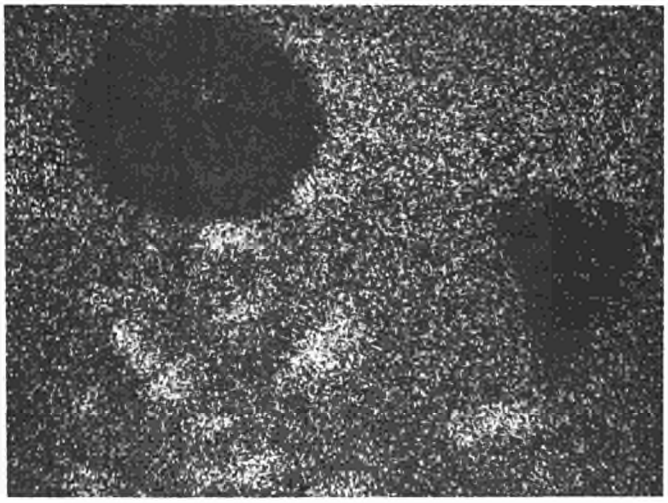


Fig. 5.2.7c
EPA-Element distribution of Si in Fig. 5.2.8a

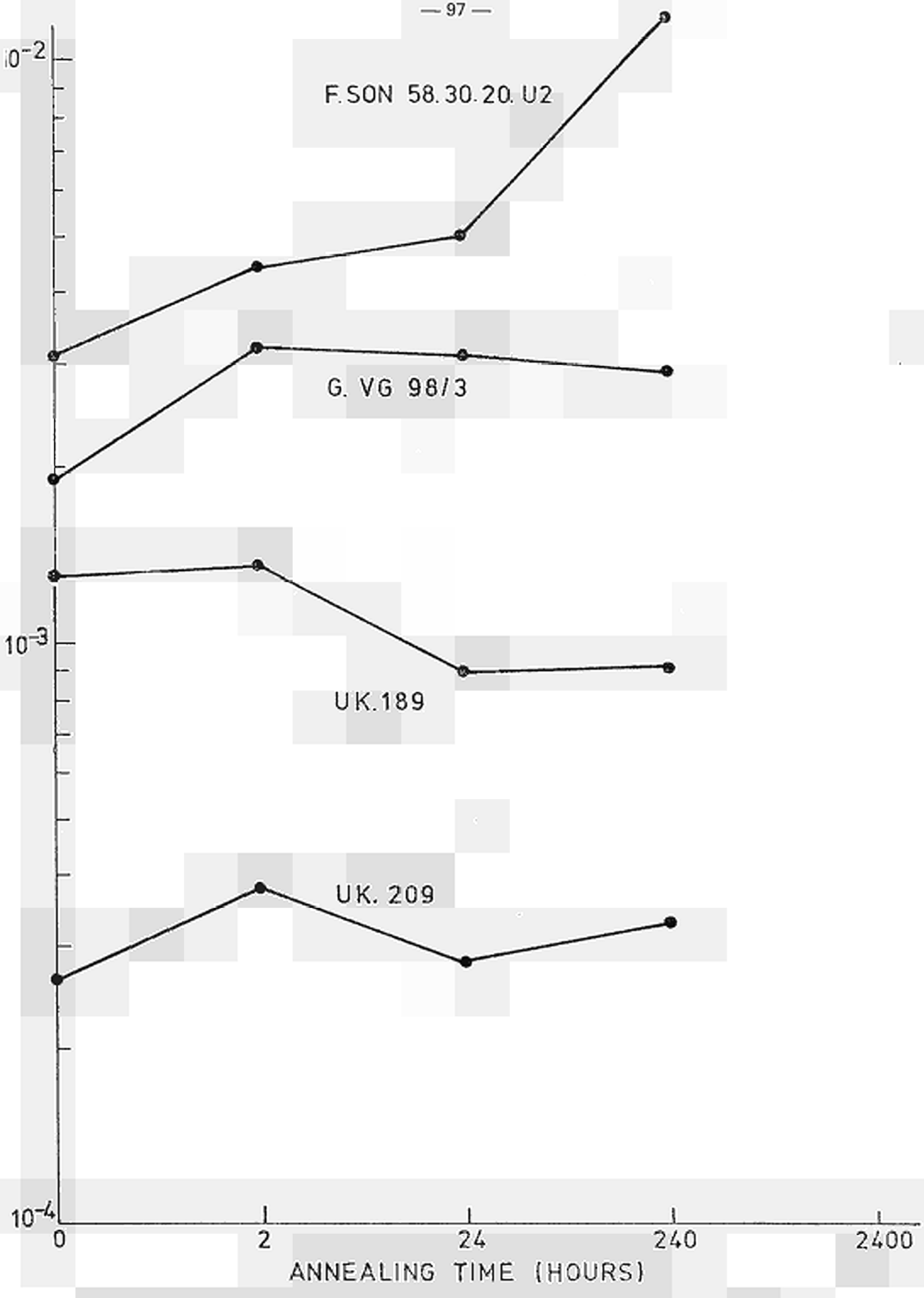


Fig.5.3.1 : LEACH RATES AFTER ANNEALING AT 800° C

6. ENGINEERED STORAGE FOR SOLIDIFIED HLW

6.1. Scope and State of the programme

The programme has been limited to two basic concepts : the water cooled and the air cooled concept, with the option of a mixed system, providing a water cooled pond for the first period of decay heat removal and air cooling for the remainder storage time. In order to obtain comparable data on cost and safety of the different storage systems, the study will be based on a set of common reference data, like construction codes, provisional design and safety criteria and common costing systems, which will be agreed upon in progress meetings of the participating firms : Nukem and Belgonucléaire. The programme is subdivided in four stages and the distribution of tasks within the four stages are given in table 6.2.1., whereas table 6.2.2. shows the time schedule.

Nukem has prepared a set of reference data, defined the properties of the solidified waste and presented the preliminary design for a storage container.

6.2. Common Reference Data and Options

6.2.1. General terms of reference

- Annual amount of spent fuel reprocessed : 1400 metric tons of heavy metals (tHM).
- Interim storage time of vitriefied HLW : 50 years.
- Mode of final disposal : geologic formations.
- Hypothetic site conditions : not yet defined.

6.2.2. Spent fuel reference data

- Type of fuel : enriched UO_2
- Origin : pressurized water reactor (PWR)
- Initial enrichment : 3,2 % U_{235}
- Cladding material : Zircaloy 4
- Average burn-up : 36.000 MWd/tHM
- Precooling time : 365 days.

6.2.3. High level liquid waste reference data

- Reprocessing technique : PUREX-process connected with a chop-leach head end.
- Acidity : 5 molar nitric acid.
- Total salt contents : 425 g/l.
- Fission products (FP) : 74,4 g/l.
- Actinides : 13,2 g/l.
- Radioactivity : 6.100 Ci/l.
- Heat generation : 30 W/l.

The heat generation versus time of the liquid waste is plotted in Fig 6.2.1.

6.2.4. Tank storage management and reference data

- Available volume per tank : 1000 m^3
- Maximum admissible total heat production per tank : 13,6 MW
- Number of tanks : 3 in action and 1 spare
- Filling sequence : subsequently 591 m^3 each in intervals of 347 days (see Fig. 6.2.2.).

With this scheme the total heat production is lower than 13,6 MW (estimated to 12,6 MW) which is the admissible value for a container of 25 cm diameter stored in a water cooled pond (see Fig. 6.2.3.). After filling the second tank the heat production in the first tank has decreased to 5,4 KW/tHM, which is below the admissible value for a free convection air cooled storage (see Fig. 6.2.3.). The solidification of the HLLW can be started approximately two years (694 days) after initial filling.

6.2.4.1. Risk of tank storage

According to the fault-tree-analysis of a hypothetical reference-system (1), the most hazardous event (failure of nearly all barriers leading to a release of radioactive material to the atmosphere via cooling tower) provides a release of about 10^4Ci with a frequency of 10^{-4} per year leading to an expectation value of 0.012 mrem per year of inhalation dose. The total expected inhalation dose taking into account all fault-tree-results amounts to a value of 0.02 mrem per year.

6.2.4.2. Costs of tank storage

The total costs for a tank storage, consisting of 4 tanks of an available volume of 1000 m^3 each, have been estimated to amount to 130 Millions DM.

6.2.5. Solidification product data

By solidification the liquid waste volume is reduced from 426 l HLLW to 73 l solidification product per metric ton of heavy metals. The characteristics of the solidification product depending on the preceding strategy are given in the table below :

Characteristics	Immediate solidification without tank storage (appropriate for water cooled storage)	After 694 days tank storage (appropriate for air cooled storage)
Fission products	434,2 g/l	434,2 g/l
Actinides	77 g/l	77 g/l
Radioactivity	$35,7 \times 10^3$ Ci/l	$15,7 \times 10^3$ Ci/l
Heat generation	180 W/l	72,3 W/l

The physical properties of the vitrification product, consisting of ~80 % borosilicate glass and ~20 % waste oxyde are :

- Density 2,9 - 3,1 g/cm^3
- Thermal conductivity 1,1 - 1,3 W/m°K
- Leach rate 5×10^{-5} g/cm^2 d
- Maximum stable temperature 900°C
- Viscosity at 900°C 4 c 10^3 Poise

6.2.6. Preliminary design data for the storage container

The shape of the solidified waste blocks has been confined to a simple cylindrical form though admitting an internal finning. As the softening point of borosilicate glasses lies between 800°C and 850°C, the maximum admissible central temperature of the blocks has been fixed at 700°C. The maximum admissible surface temperature for the container is fixed at approximately 60°C for the water and 450°C for the air cooled concept. The geometry of the container (see Fig. 6.2.4.) has been defined as follows :

- Outside diameter 267 mm
- Length 1750 mm
- Free volume ca. 73 l

6.3. Programme of work for 1978

Stage I of the programme "the definition of the two basic storage schemes" is, as indicated in table 6.2.2., in progress since October 1977 and will be terminated in April 1978.

The target of Stage I is to establish the design data for heat removal systems, storage volume, layout of storage and handling systems, draft schemes for electrical and other supply systems, general flow sheets, preliminary common safety criteria for the conceptual design study and requirements for the risk analysis and similar basic design information.

The remainder of 1978 will be devoted to state II which deals with the assessment of common safety criteria and design rules applicable within the frame of the design and evaluation study of this programme.

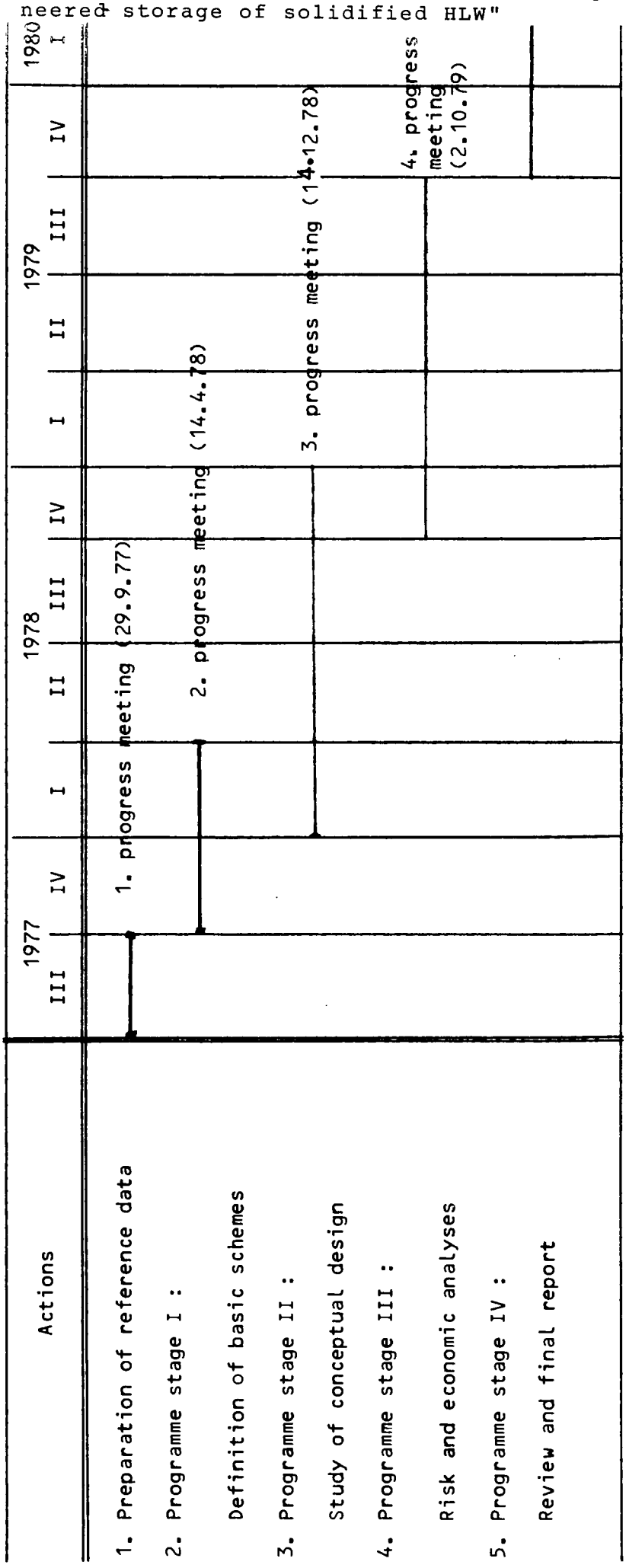
6.4. Reference

(1) SRA-5, H. J. Wingender, Systemstudie : Radioaktive Abfälle in der Bundesrepublik Deutschland (KWA 1214).

Table 6.2.1. : Distribution of tasks for programme sheet 6 Engineered storage of solidified

	NUKEM	BELGONUCLEAIRE
Stage I	1. Preparation of reference data	
	2. Definition of properties of solidified waste	
	3. Definition of preliminary container I	
	4. Basic scheme of air cooled system	1. Basic scheme of water cooled system
Stage II	5. Assessment of common safety criteria	
	6. Design of definite container II	
	7. Conceptual design of air cooled system	2. Conceptual design of water cooled system
	8. Design of handling and monitoring systems	3. Design of handling and monitoring systems
Stage III	9. (Conceptual design of air cooled part of mixed storage if adequate)	4. (Conceptual design of water cooled part of mixed storage if adequate)
		5. Common costing data
	10. Cost estimate for air cooled storage (and possibly mixed)	6. Cost estimate for water cooled storage (and possibly mixed)
	11. Risk analysis	7. Economic analysis
Stage IV	Joint final report	

Table 6.2.2. : Time schedule for programme sheet 6 "Engineered storage of solidified HLW"



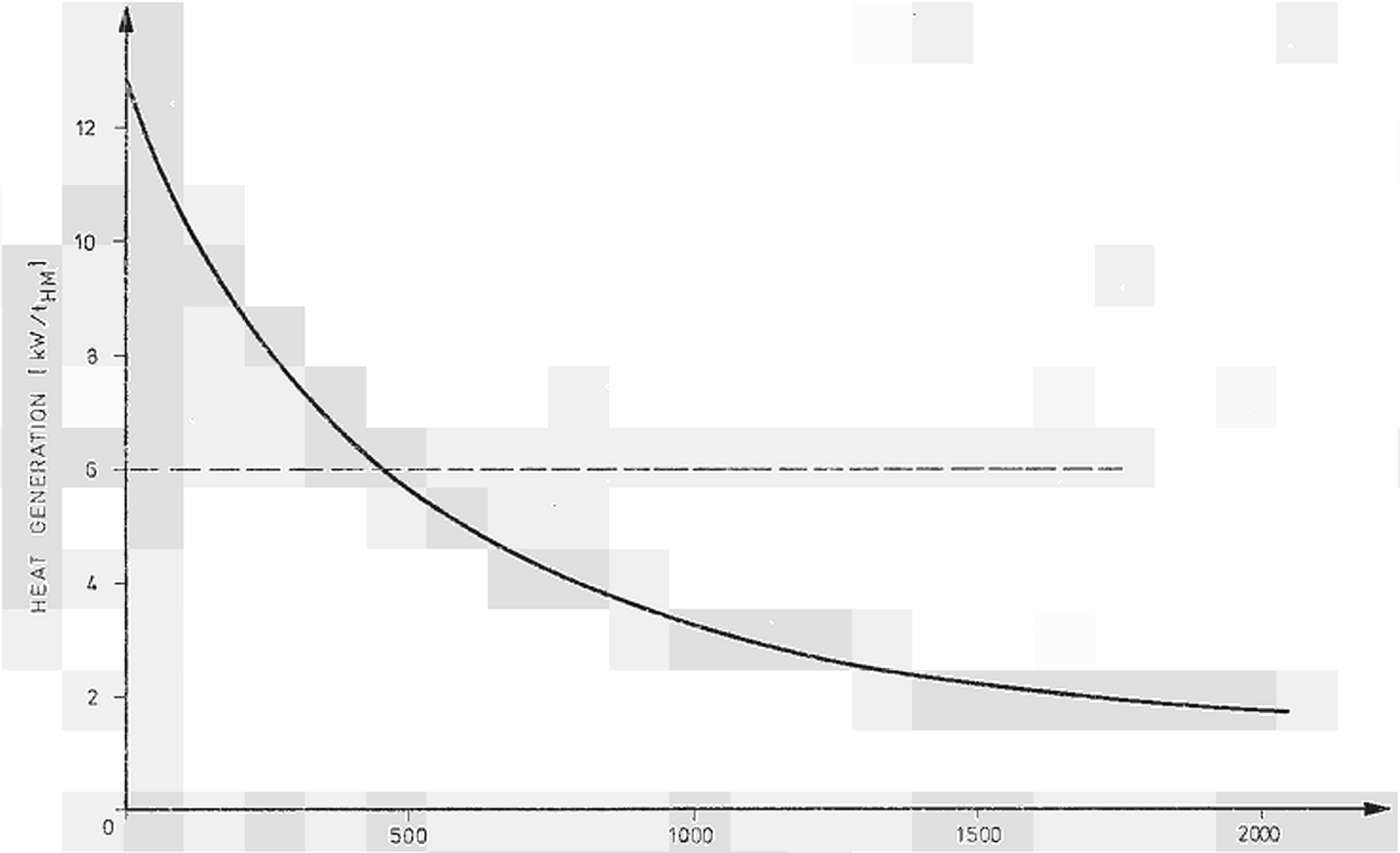


Fig. 6.2.1 : Heat Generation of High Level Radioactive Waste (in kw/t....)

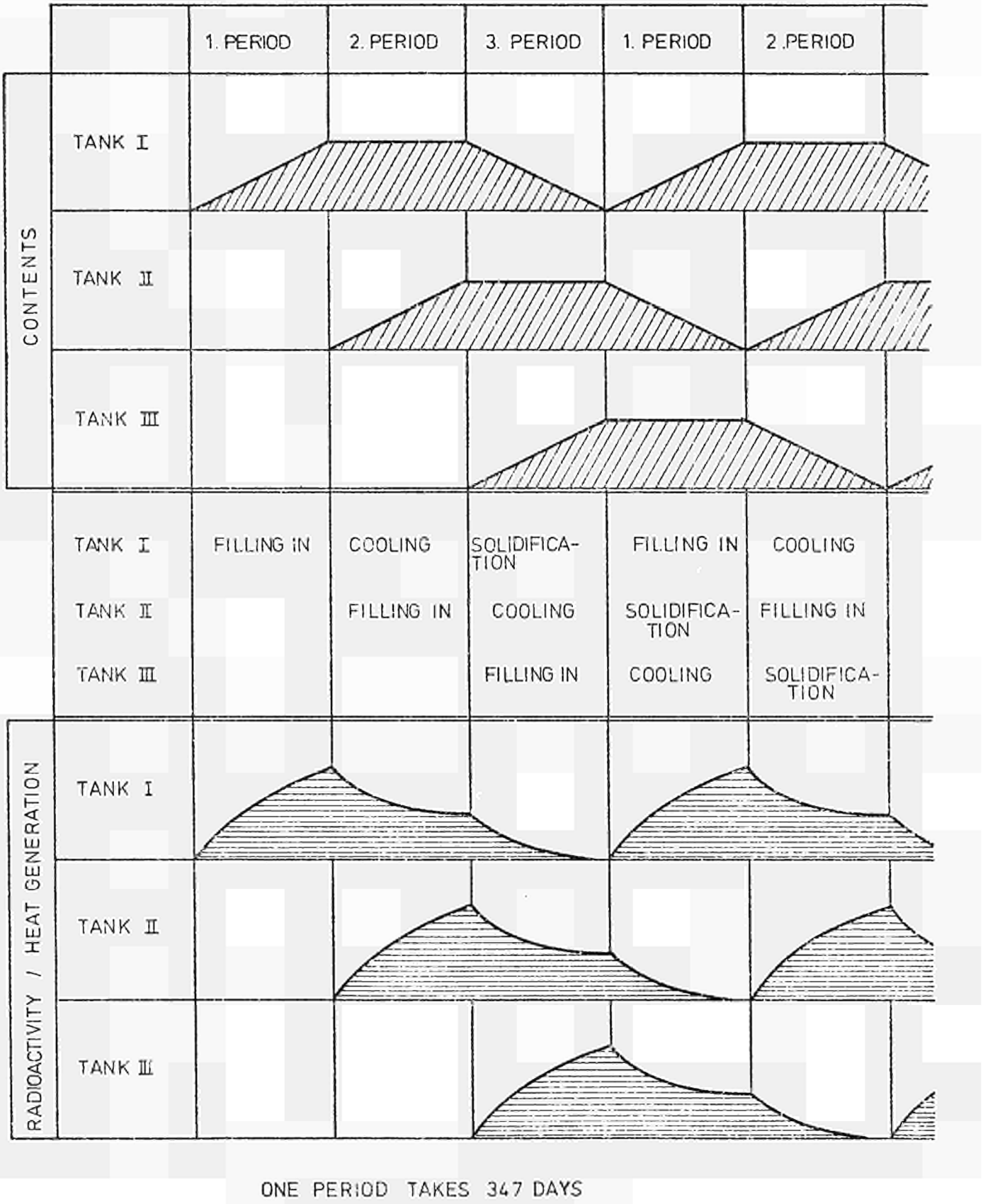


Fig. 6.2.2 : Graphical description of the proposed tank-storage concept for solidification in the shortest possible time (qualitative)

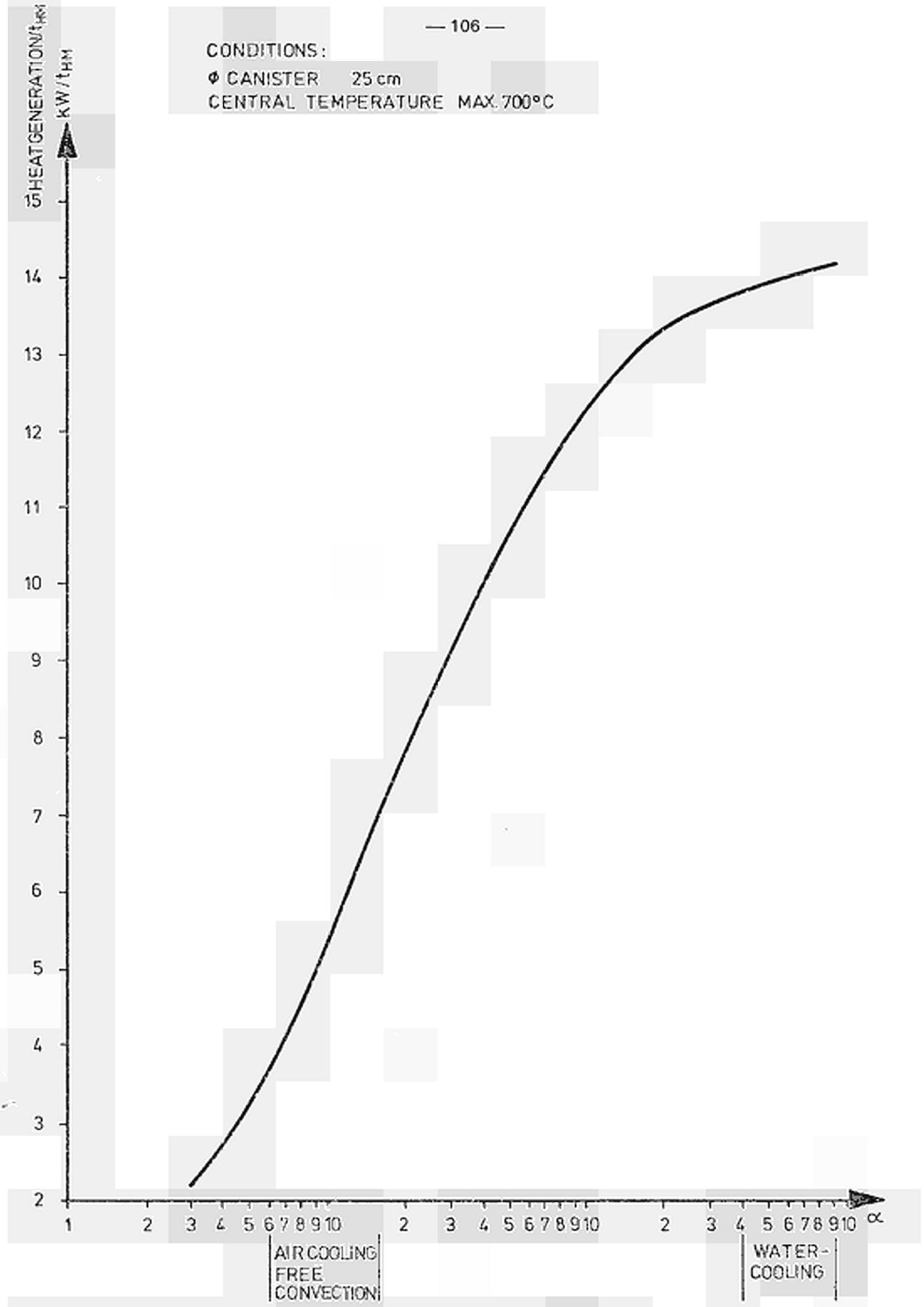


Fig. 6.2.3 : Heat removal in block storage (in kW/t_{HM})

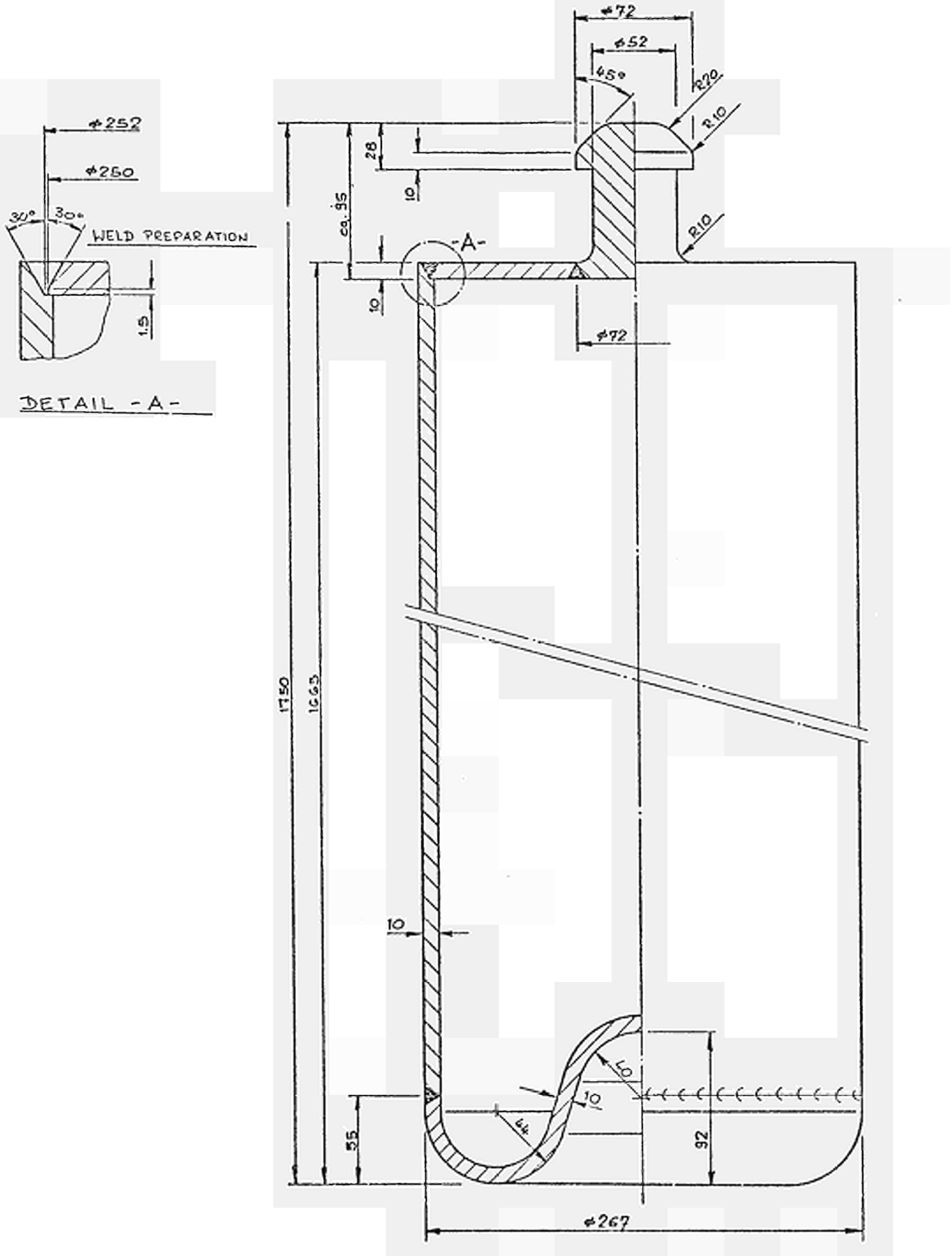


Fig. 6.2.4 : 1st Design of the HLW-canister

7. DISPOSAL OF RADIOACTIVE WASTE IN GEOLOGICAL FORMATIONS

7.1. Introduction

The objective of this action is to investigate the possibilities of isolating toxic radioactive wastes into appropriate geological formations in such a way that the public health and safety will not be endangered during the period of time required for their natural radioactive decay to innocuous level.

Typical types of geologic formations were distributed among member States for investigations, each country concentrating its study on one rock type taking into account :

- a possible past involvement with a specific type of rock (like Germany with salt)
- the underground characterics of the national territory (like Belgium with clay)
- the necessity to save money in avoiding non necessary duplication of high cost experiments like deep drilling.

As a result, clay formations are investigated by Belgium and Italy, cristalline rocks (granite) by France and UK, and salt domes by the Federal Republic of Germany and the Netherlands ; supporting studies are going on in Denmark and Ireland. This does not imply a national commitment to any type of disposal method or rock type at this stage, with the exception of Germany.

The goal of the E.C. programme is actually to make possible such definitive commitments in the future by making available all the required information at the proper time. Its main topics are :

- general survey within the E.C. countries of potential areas for radio-active waste geologic disposal ; such a survey at Community level was initiated in 1976 on the basis of the existing national information. A Catalogue, using criteria agreed in common, will be issued at the end of the present programme (1979). It will provide an overall view of the geological formations of potential interest for radioactive waste disposal in the Member States of the European Communities ;
- determination of site characteristics such as geologic, geophysic and hydrogeologic data related to the repository integrity ;
- determination of the ability of the formation and of the rock to meet the integrity criteria ; establishment of the site integrity criteria ; experimental confirmation on lab scale or in situ and corresponding

data modelling ;

- repository storage capacity evaluaions and design studies ;

- development and/or choice of adequate technical systems and equipment for repository construction ;

- evaluation of waste isolation properties of the site barrier by barrier (waste conditioning and container ; repository features ; migration of radio-nuclides to biosphere) and overall safety evaluation by means of risk analysis and modelling.

Table 1 gives an overview of the programme contents.

Table 1 : Underground disposal (period 1976 - 1979)

* completed
\+ in progess
= scheduled

	SALT		CLAY		CRIST ROCK			
	GfK D	ECN NL	CEN B	CNEN I	CEA F	UKAEA GB	RISØ DK	AEC IRE
1. Catalogue of formations of interest	*	*	*	*	*	*	*	*
2. Geologic studies and associated confirmation works of research sites								
- deep core drillings	*	=	*	=	=	=		
- hydrogeology		=	+	+	+	+		
- other studies		+	+	+	+	+		
3. Technical support activities								
- heat transfer, thermal analysis and associated studies	+		+	+	+	+		
- rock mechanics and other properties	+		+	+	+	+		
- waste rocks interaction	+		+	+		+		
4. Engineering and associated studies								
- conceptual studies of repository	+	+	+	+		+		
- waste canister design	+		+	=				
- transportation technology	+		+	=				
- backtilling and plugging			+	=		=		
5. Hazard analysis and modelling of the migration of radionuclides to the biosphere including labs and in situ experiments		+	+	+	+	+	+	
6. Safety report and licensing	+		+	=				
7. Start construction of the shaft	* 1974		= 1979					

7.2. Disposal of Radioactive Waste in Salt Dome (RFK)

7.2.1. Objectives

Construction in the next years of an experimental site in the Asse mine to assess the possibility for the disposal of high level waste in a salt dome.

In 1977, works essentially consisted of :

- Simulating the influence of heat generated by radioactive waste in the properties of the surrounding salt rock (temperature distribution, galeries and boreholes convergence, migration of release of enclosed brine droplet, modification of the salt rock rheological properties, determination of absolute stresses in the formation ; works have been carried out in laboratory and in situ together with the development of calculation procedures for the investigation of these phenomena ;

- Developing technical systems and equipment for transportation, emplacement and retrievability of waste containers and for monitoring of the repository ;

- Investigation of some aspects of waste isolation and repository safety (corrosion of borehole casing and canister materials, consequences of an inrush of water into the underground workings).

7.2.2. Determination of the ability of the formation and of the rock to satisfy the integrity criteria

7.2.2.1. Experiments to determine the temperature distribution and the convergence in lined and unlined boreholes

a) Simulation experiment in older halite with unlined boreholes

To supplement the temperature experiments carried out so far in younger halite, an additional test in older halite is necessary, since older halite possess different characteristics in respect of heat conductivities, rheological parameters and behaviour of the brine inclusions.

The experiment includes measurement of the temperature fields, the uncased borehole convergence, the borehole climate and the corrosion of the materials. The relative humidity and temperature of the air in the borehole are of interest, because they influence the corrosion of the materials. The rock temperature and convergence will be recorded to determine the behaviour of the Older Halite regarding elevated temperatures. The data will be compared with the results

of theoretical calculations. It is expected that brine droplets which are included in the salt will migrate towards the heating source because of the high temperature gradient in the near surrounding of the borehole. Assuming a temperature of about 150°C (100°C < t < 200°C) in the borehole the brine will vaporize and induce an increase of pressure.

For the preliminary test a horizontal borehole in the vicinity of a wall corner has been used on the 775 m level in the Asse salt mine. Small boreholes perpendicular to the main borehole were drilled and temperature gauges installed. The air in the gas-tight borehole is periodically blown through a cooling trap to measure the amount of vapour after condensation in a calibrated measuring tube. The condensate is vapourized again and pumped back into the borehole in order to avoid a disturbance of the climate during the duration of the experiment. A pressure gauge is installed to measure the gas pressure in the borehole. As the temperature in the borehole is known, it is possible to calculate the relative humidity.

After the preliminary test has been finished, the main experiment will be started using a vertical borehole in the Older Halite(temperature test field 4, Fig. 2.2.1.). Rock deformation caused by temperature will be recorded by extensometers. All data will again be compared with theoretical calculations. In order to measure the closure of the borehole caused by elevated temperatures a system of five in-line measuring probes will be installed.

b) Simulation experiment with cased borehole

This experiment is designed to provide information on the compressive load to which a quasi-rigid casing in a converging storage borehole is subjected. On the other hand, this is of direct interest as regards possible retrievability of the emplaced high-level glass cylinders ; on the other hand, the pressure to which the casing is subjected is an extremely important parameter for checking out the rock-mechanics computer programme.

The experiment will be run in two steps. (1) A single borehole will be cased and instrumented to measure the casing parameters (strain and temperature) depending on the heating power and time. (2) Another similarly cased borehole will be surrounded by 6 equidistant boreholes forming a hexagon. These 7 thermal sources will simulate the influence of a temperature field on a cased borehole. The concept of the electrical heaters is rather flexible and can be used for various heater forms.

A testing facility was constructed consisting of a heater of this kind placed in a sleeve termally insulated. The temperatures on the heater were measured. The temperature gradient was small enough to produce a homogeneous temperature distribution on the sleeve. Variations being less than 1 % were observed.

For the measurement of the pressure load on the borehole casing coming from the termally induced borehole closure, special pressure pillows welded on the surface of the casing and filled with mercury will be used. The load will be recorded by high temperature pressure gauges, provided that the wall of the borehole is smooth enough. In order to prove this smoothness a borehole measuring probe has been developed, which allows to measure the surface relief and two orthogonal diameters of the borehole. Up to now, two boreholes have been measured in such a manner. The test measurements showed that the deviation from the circular cross section is within ± 2 mm and no significant variation of the surface roughness of the walls of the boreholes were observed, due to heating and closure.

c) Development of a standard probe for systematic investigations on thermally induced borehole closure

The standard probe, which consists basically of a heater and an integrated device for the in-line measurement of borehole convergence, is intended, in a series of identical tests at various sites in the Asse mine, to provide information concerning the dependence of the heat-induced borehole convergence on the rock stress conditions and the characteristics of the materials in the surrounding massifs. With these results, it would be possible to check the extent to which the appropriate computer programmes can take into account various rock stresses and material characteristics. The experiments with the standard probe will thus be an important adjunct to the simulation tests. (see para. 7.2.2.1. a) and b)).

The constructions of the standard-probe for the heating of boreholes and the measurement of closure rates has been continued. The design of the probe itself, consisting of the central heater body and two shorter non-heated parts is almost finished. The probe is instrumented with 5 ring shaped electrodes for the in line measurement of the borehole closure. (see Fig. 7.2.2.).

For testing the outer ring electrodes which have to function-even during the closure, a special apparatus has been constructed. First tests showed satisfactory behavour of the ring electrodes for the standard probe.

The data logging and control electronics and the heater powersupply unit were ordered. The microprocessor controlled electronics unit includes the usual function units of a modern data acquisition system. The use of the microprocessor will provide an easy way to connect the standard probe to the computer system which will be installed in the Asse mine.

d) Measurement of convergences of galleries and boreholes in connection with temperature experiments already carried out or to be carried out.

These measurements supplement the measurements of borehole convergence (or of the convergence pressure on the casing) performed in the simulation tests. To ascertain that part of the early history in which no influence was exercised by heat, such measurement must be carried out well before the heat experiments begin.

First measurements of deformation have been carried out in younger halite (temperature test-field 3). The main results are following below :

- A thermal source (5 m long, 0,2 m diameter, 9 kW heating power) did not cause deformations in a neighbouring borehole (7,5 m horizontal distance, 320 mm diameter) during a heating period of 90 days

- In the heated borehole an accelerated borehole-convergence was noticed only in a distance of up to 4,5 m above the heating elements

- The maximum borehole-convergence correlated with the central level of the heating source. The convergence measured 99 mm after a heating period of 90 days. (Fig. 7.2.3.).

- Under the specified testing conditions the heating element was partly surrounded by the salt-formation after a period of about 90 days. The heating element was installed in an unprotected borehole without any casings.

- A second thermal source with the same dimensions could be installed above the first one in the same borehole without any problems even 90 days after the installation of the first one.

- The emission of heat from the temperature-test-field No. 3 did not cause any additional deformations in the workings or the pillars of the higher level of the testing-site.

Preliminarly investigations were done concerning planning and instrumentation for the measurement of

rock-deformation in the older halite by heating tests. Correlating measurements with wire extensometers and rod-extensometers showed, that the wire-extensometers, favoured financially and technically, should be restricted in their use to a depth of 20 m in the formation. In extended depths rod-extensometers should only be used to assure reproducible results.

c) Determination of absolute stresses

The knowledge of the absolute stresses of the rock formation is necessary for the judgment of the stability of underground cavities, and the formulation of realistic boundary conditions for rock mechanical model calculations. It is also necessary to know the rock parameters and the special in situ conditions.

The compensation method, has been checked ; it is more advantageous than other methods measuring the redeformation when the Young's modulus and Poisson's ratio are known and the deformation is completely reversible after overboring the borehole because a knowledge of the Young's modulus and the Poisson's ratio is not necessary. It is based on an artificial redeformation of the rock samples to produce the original stress values in the rock formation.

It can be expected that the distributions of stress in salt rocks will only be determined qualitatively by techniques based on the deformation measurements. However, it will probably be possible to measure absolute stresses of salt rock using a salt gauge based on the compensation method. This technique will therefore be applied at first.

7.2.2.2. Determination of rheological properties of Asse rock salt at elevated temperature

The underground disposal of highly radioactive and heat producing waste results in additional creep phenomena in the rock formation which are caused by the superposition of the temperature field and temperature induced stress. These phenomena depend strongly on the mechanical properties, especially on the long term creep characteristics of the salt rock in the vicinity of the site. By means of uni- and multi-axial stress and deformation tests on Na-2 and Na-3 at temperatures up to about 400°C, the aim of the study is to establish the basis for formulating creep laws.

A laboratory test programme has been started by GSF and the Bundesanstalt für Geowissenschaften und Rohstoffe (BGR) Hannover to describe these phenomena in laboratory. Three series of rock salt cores have been taken from different underground places of the Asse anticline :

The first cores come from an area where a temperature test field will be installed. The second series comes from a region which is deep in the center of the anticline. Series 3 comes from that area, where the old galleries are situated within the section of Younger Halite. It has been selected to yield representative results in comparison with the first two series.

a) The elastic dynamic parameters of the same rock have been determined by the ultrasonic pulse method. The samples have been tested in two directions (parallel and perpendicular to the cylinder axis) ; the maximum error can be estimated to be between 1 and 2 %, depending on the type of waveform and direction of pulse transmission.

   Comparing the data based on calculations with the corresponding laboratory values of different Asse salts rocks, it can be seen, that there are very low differences between them. (See results for older halite on Fig. 7.2.4.). It can be concluded, that near face of old and spalled underground rooms, openings or cavities the in situ rock salt has a looser structure, because of the micro-cracks of the rock formation. The velocities of ultrasonic waves are therefore much less than they are in rocks not spalled.

b) The uniaxial creep experiments have been carried out using cylindric specimens of a diameter of 100 mm and a length of 250 mm. At first, the uniaxial compressive strengths of the samples σ_c have been determined. The constant loading rate used was $\dot{\sigma}$ = 1,0 MN/m^2. min. The results are as following :

   Series No 1 (Na 2 U) : σ_c = 29,5 (MN/m^2)

   Series No 2 (Na 2) : σ_c = 26,2 (MN/m^2)

   Series No 3 (Na 3) : σ_c = 27,6 (MN/m^2)

   These data will be currently verified by parallel compression tests. The creep tests have been carried out at stress levels between 4 and 18 (MN/m^2). The test temperatures were in the range of room temperature and 573°K (300°C). Preferably, the temperatures of 373, 423, 473 and 573°K have been used ; 3 tests of 40 tests started have already been finished. The preliminary results, currently plotted in ε - t - diagrams, show the enormous influence of temperature on the creep behaviour and the creep rate. A further data analysis is in preparation to find out the functional connection between the apparent creep rate $\dot{\varepsilon}$ and the parameters ε, σ, t, and T.

c) First triaxial creep tests of 17 cubes have been performed with the triaxial test apparatus of the BGR at multiaxial stress levels ($\sigma_1 \neq \sigma_2 \neq \sigma_3$) using cubes

having sides of a length of 110 or 120 mm respectively. The results of these tests are analysed to gain experiences for further investigations.

7.2.2.3. Improvement of existing computer programmes for the calculation of temperatures in the glass and in the salt

This improvement has been made for individual disposal hole arrangements as well as in the salt formation outside the disposal area and the overlying formations. A set of various codes for both unit cell calculations and investigations of the global temperature distributions throughout the disposal field and geology formation area available now.

a) A special code for the evaluation of temperature field experiments has been written and was used to compare the experimental results from two field experiments performed in the Asse salt mine with theoretical predictions (see section 7.2.2.1. d)). The results of the calculations, which were carried out for the temperature experiment 3, are essentially too high. The temperature curves for the two points a and in the center of the heater were compared. Point a is situated at the borehole edge and point b in a distance of 1 m from the borehole. The examinations show that the discrepancies can be interpreted as following :

   - In a less degree they are based on differences in describing the geometry (width of the air gap and the length of the heater) and the heat generation (slightly decreasing heater power during the experiment, power interruptions, heat dissipation from the Chrome-Nickel-lead- in wires).

   - In a high degree the differences must be due to the convective vertical heat exchange in the borehole. This was the result of an additionally calculation with a modified version of the code ASTEFE. Here the convective heat exchange was simulated by a fictitious heater (length 1.5 m and a heat power of 29 % of the total heat power) which is placed above the heater.

   - For point b the results of the calculation then almost agree with the test readings, only a small further increase of the convective heat exchange portion might bring a full agreement. So it can be concluded, that at least 20 % of the heat has escaped from the heater region due to convection.

b) As a further result of this programme a tested three-dimensional computer code in Cartesian coordinates is developed, which allows the calculation of the transient

temperature rises in a multiform salt formation and its surrounding rock formations after disposal of high-level radioactive waste. With this code CUB3DG calculations were made for some typical geometries of the salt formation. The considered cases are characterized as follows :

- Below a rocky surface layer of 280 m there may exist a salt formation with an approximately quadratic cross section

- The disposal area may be positioned at the vertical center line of the salt formation in a depth of 800 m ; its also quadratic cross section may have an area of 0.5 km^2 and a height of 50 m. A disposal area of this size can contain the solidified waste of a 20 year production of a 1400 t/y reprocessing plant

- The throughout the disposal area smeared heat dissipation of the waste will result in a specific power level of 1.14 W/m^3, under the assumption that the age of the waste prior to burial will be 10 years.

- The horizontal distance between the disposal area and the rock formation will be assumed as approximately 300 m (case A), 200 m (case B) and 100 m (case C).

The results of the thermal analysis indicate, that there is no influence on the maximum temperature in the disposal area (215°C after appr. 50 y) if the horizontal distance between mine area and rock formation changes from 300 m to 100 m. By decreasing the distance the horizontal heat resistance will increase and this will cause an accumulation of heat between disposal area and rock formation.

If the computed temperatures for flank distances of 100 m and 300 m are compared, the results show, that there is nowhere in the salt formation an additional temperature rise of more than 6 K. Only for cases with extremely small distances between disposal area and the rock formation or in the case the rock formations have a very low heat conductivity, greater temperature effects in the salt formation would be possible.

The highest rock temperatures arise at the boundary of the rock formation at the level of the disposal area. The temperature at the boundary of the rock formation rises to 62°C (after 150 y) if the distance between disposal area and the rocks is 100 m and to 41°C (after 550 y) in the case of 300 m. In these two cases the horizontal mean temperature gradient in the first 100 m of the rock formation will be at

its maximum 0.23 K/m and 0.04 K/m respectively ; the smaller value is consequently less than the vertical geothermal gradient of 0.054 K/m, which was used in these calculations.

7.2.2.4. Development of computer codes for calculation of the stability of single pillars and the whole disposal field at elevated temperature

The basis of reliable geomechanical calculations must be the knowledge of the materials behaviour of rock salts depending on stress, temperature and time. Therefore, it is very necessary to have a complete catalogue of data containing all materials parameters of the site of the repository. These parameters of a comprehensive constitutive law must be determined during short-term and long-term measurements of a lot of salt rock samples in situ and in laboratory. The development of numerical methods for rock mechanical stability calculations has been started in a twofold way : it was investigated whether and which appropriate computer codes are already existing and a programme system considering the special thermomechanical and long-term behaviour of salt rocks was developed.

7.2.2.5. Migration and release of enclosed brine droplet in older halite

The considerable temperature gradients in the immediate vicinity of the borehole can cause alkaline droplet inclusions to move in the direction of the borehole, thus resulting in an increase in the occurrence of borehole humidity. This would produce undesirable side-effects, e.g., decrepitation of salt crystals and increased corrosion.

a) Determination of the water components of older and younger halite.

Rock salt formations may contain up to 1 % of volume of water. This water can be either water of crystallization or thin water film on the boundary surface of the crystals or enclosed brine droplets. In order to determine the different water components in the Older and Younger Halite four different methods are used :

- Vacuum drying at a temperature between 105 and 180°C.

  The Younger Halite had an average loss of weight of 0,01 % whereas there was no loss in weight of the Older Halite.

- Infrared drying

  The generated temperature in the salt was about 120°C. After about 72 hours of drying the average loss in weight of the Younger Halite was about 0,015 % whereas the loss of weight of the Older Halite was less than 0,01 %.

- Thermogravimetry

  Up to now, Older and Younger Halite of the Asse Salt Mine from depths of 775 and 975 m were analysed. With a heating velocity of 10°C/min the Younger Halite showed a sudden loss of weight at a temperature between 270 and 280°C, being caused by the release of the water content. When rising the heating velocity to 20°C/min the signal appeared at about 290°C, when reducing the velocity to 0,2°C/min the signal appeared at 260°C. The average water content released at temperature between 270 and 280°C was between 0,06 and 0,1 %. In order to determine the inhomogeneity of the water distribution within the rock salt, samples of about 3 grams were taken from different parts of a larger block of rock salt. The highest water content was 0,16 % and the lowest was 0,03 %.

  The thermoanalytic investigation of the Older Halite was the same like that of the Younger Halite. But the results were different. The Older Halite of the 775 m-level had a water release of 0,04 % at a temperature of 290°C with a heating velocity of 10°C/min. The samples of the cavity showed a water release of 0,02 % at a temperature of 450°C.

  These results demonstrate that there are different water components and different physical and chemical conditions. Besides their characteristic sudden loss of weight at temperatures of about 280 or 450°C, respectively, all samples of Younger and Older Halite showed a further loss in weight beginning at a temperature of about 600°C. Whether this last loss of weight is caused by another water release or by a chemical conversion of some minor halite impurities could not be proved yet.
  Above the temperature of 620°C the halite begins to sublime.

- Karl-Fischertitration

  Up to now the Karl-Fischer-titration indicates that at least one part of the predictable water content of the salt depends on the fineness of grinding. This demonstrates that there must exist different water components and different physical and chemical conditions. Further investigations must be performed to distinguish these components.

b) Determination of the diffusion and release of the water at constant temperatures

- In order to determine diffusion and release of the water at constant temperatures, cylindric rock salt samples of 15 cm length and 5 or 7 cm diameter were put into a drying oven at a temperatyre of 200°C. After drying samples were cut into thin disks of a thickness of 5 mm and the water content of the salt was determined by termogravimetry. A concentration gradient of water was found to exist in their longitudinal direction. Samples of Older and Younger Halite will be put into the drying oven at temperatures between 50 and 400°C in order to determine the speed of release and the diffusion at different constant temperatures.

- In order to investigate the thermodiffusion of the water content caused by the temperature gradient, cylindric samples were placed between two heating plates with temperatures of 150 and 200°C respectively which caused a thermal gradient of 3,3°C/cm. Brass tubes were shrinked around the samples, the samples cut into thin disks and the water content analysed.

c) Preparation of an in situ confirmation test.

On the 775-m level of the Asse Salt Mine a temperature-test field was prepared which consists of a horizontal borehole of 28 cm diameter. In order to measure the temperature in the salt 25 thermocouples were put into the salt around the borehole. Before starting the heating. The water content of this Older Halite was determined to be about 0,04 %. After about half a year of heating with 4000 watts the test will be finished and series of salt samples of different parts around the borehole will be taken and analysed.

7.2.3. Technical systems and equipment

This set of projects covers the development, procurement and installation of all technical facilities and instruments necessary for the transport, emplacement and retrievability of the high-level waste cylinders, together with the procurement of the measuring instruments to be used for monitoring in the planned experimental disposal.

7.2.3.1. Development of a shielding container for HLW glass blocks

It has been begun to develop the shielding container, to meet the requirements of the Asse mine with respect to weight (9,8 t) and diameter (1,2 m) and the conditions demanded by the supervisory licensing authorities.

a) Shielding

The calculations carried out with the computer code SABINE showed that the optimum cylindrical shielding is a combination of 22 cm of lead and 12 cm of polyethylene with 2 % or 10 % of B_4C. But a more accurate transport calculation is still necessary to consider the neutron shielding aspect. In order to avoid the difficulties of the one-dimensional SABINE code, a threedimensional Monte Carlo programme (SAM-CE) has been used for neutron calculation.

- Neutron shielding calculations : For analytical calculations being sufficient to compute the gamma radiation shielding , two computer programmes are available. The calculations have been carried out for various shielding situations because, besides the normal transport situation (22 cm Pb, 12 cm PE with 2 % B_4C), also accident situations with reduced shielding thicknesses must be taken into account. The results show that especially polyethylene produces an attenuation effect in the area of the lateral shielding shell of the same order as the layers of steel in the top and bottom lids. The highest dose rate must be expected to occur at the bottom of the container.

- Gamma radiation calculations : For calculating the gamma shielding a plane shielding layer has been assumed because in the analytical programmes only plane layers are allowed. The flux density of the unscattered radiation behind a plane layer is always higher than behind a curved layer. The gamma dose rates for the areas of the top and bottom lids of the transport container can practically be neglected compared with the neutron dose rates.

- A shielding container model on a 1:4 scale will be used for experiments with gamma and neutron sources in order to test the accuracy of the methods of computation and to optimize and select alternatives for the transport container, the canister grab and the borehole slide systems.

b) Canister grab systems

For a preliminary storage concept is has been first planned, to install the canister grab in the transport container. The release mechanism actuated by a lifting magnet is firmly attached to a wire coupled to the grab for unloading the container and for recovery (Fig. 7.2.5.). In anycase, the recovery of the canister including the rupture of the lift wire will be guaranteed.

7.2.3.2. Development and purchasing of technical facilities and systems for transport, storage and recovery of HLW glass cylinders

In addition to the existing design concept of the technical facilities for storage and recovery of high level glass cylinders, alternatives have been designed. The final decision about the high level waste storage concept will be taken, after the experiments with the scale model container, the simulation experiments in the laboratory and in situ will be evaluated, in accordance with other criteria (handling safety,adaptability to other storage systems, height needed for installation, and feasibility).

The canister grab, the canisters and the borehole casing (Fig. 7.2.6.) meanwhile developed are now being tested in laboratory and in situ.

7.2.3.3. Development, purchasing and assembly of monitoring measuring instruments

For the experiments to be realised in the Asse mine, especially for the planned simulation experiments (section 7.2.2.1 a) and b), the numerous different signals and data have to be recorded and processed. For these purposes a mini-computer will be used. A computer processing and control system will also be able to control the experiments. Because of the high dust concentration mechanically operated devices like magnetic discs or tape units cannot be installed in the mine. To overcome this problem , a data transfer line will connect a central unit installed in the mine with another minicomputer installed outside (Fig. 7.2.7.). The components of this system were ordered.

7.2.4. Waste isolation and repository safety

7.2.4.1. Investigations on the corrosion of borehole casing and waste canister materials in salt at elevated temperature

High-active glasses should be stored temporarily in the Asse salt mine. In connection with this experiment the retrievability of the glass blocks must be guaranteed. For this reason, the boreholes in the Asse salt mine should be equipped with a stainless steel casing.

a) Using data from the literature, a selection of materials has been undertaken on the basis of the mechanical requirements, weldability and corrosion resistance in the media to be expected. This preliminary selection was confirmed by answers given by well-

known steel producers to queries.

Only for the container material, which is mainly attacked by demineralized water during pool storage, a high-alloyed austenitic stainless steel would be sufficient. For corrosion tests the following materials are provided (according to DIN 17007) :

1.1	X10 CrNiTi 189 (≙ 321)	1.4541
1.2	X3 CrNiMoN 17135 (≙ 3172)	1.4439
1.3	X2 NiCrMo 25205	1.4539
1.4	X3 CrNiTi 3220 (Incoloy 800 H)	1.4558
1.5	X8 CrNiMoNb 1616	1.4981
1.6	NiCr21Mo (Incoloy 825)	2.4858
1.7	LC-NiCr 15 Fe (Inconel 600)	2.4815
1.8	Hastelloy CA (68 Ni, 16 Cr, 16 Mo)	2.4610

Concerning the casing material for the test storage in the salt formation, probably nickel alloys must be preferred, as in this case a corrosion attack by wet rock salt and even salt solution can be expected at increased temperatures. The corrosion tests refer to the following materials :

2.1	X2 CrNiMoN 225	1.4462
2.2	NiCr21Mo (Incoloy 825)	2.4858
2.3	NiCr22Mo9Nb (Inconel 625)	-
2.4	Hastelloy C4 (68Ni, 16Cr, 16Mo)	2.4610

However, an exact statement can only be given after evaluation of the tests.

b) Programme for corrosion tests

As generally no adequate values can be drawn from litterature (material leaflets, tables of steel producers) for the corrosion mediums mentioned, it is necessary to make tests with real mediums and conditions. The test programme is shown on Fig. 7.2.8.

7.2.4.2 Development of calculation procedures for the investigation of process which could be expected after an inrush of water or brine into the underground workings

This development is made in connection with heat generation of the HWL wastes (conversion, material exchange).

In order to have available quantitative description of the physical and chemical events in the case of a hypothetical inrush of water into a salt repository, first assumptions were developed concerning the Asse Salt Mine as a model. The physical and chemical conditions of the originating solutions were determined. Some diffusion coefficients of radioactive nuclides were measured within highly saturated brines. The self diffusion coefficients were determined by the open-end-capillary method. The

diffusion coefficients of Cs-134 within saturated NaCl-solution are determined to be 7,54. 10^{-6} cm^2/sec, within brine sample from the Asse Salt Mine to be 9,7. 10^{-5} cm^2/sec.

Field experiments within flooded salt mines were continued. It could be shown that within the shafts after some time several water-layers are formed in which temperature, specific density and salt contents increase from the surface downwards. Within such water layers, which may have a thickness of some to some hundred meters, the vertical thermogradients have an order of magnitude of 1.10^{-3}°C/m. In most cases the density, the temperature and the chemical composition are rather homogeneous inside such layers. From one layer to the next one the physical and chemical properties of the solutions change very rapidly. The transition zones have normally a thickness between 0,2 to 3 m with a thermal gradient between 0,5 to 50°C/m.

The aim of these field investigations is to have a better understanding for the development of these boundary layers and of the role which they could play if radionuclides would disseminate within the water. It can be assumed that inside these layers very slow convection streams are existing which collide at the boundaries. This effect may cause the very sharp boundary layers. It can be easily shown by experiments with small laboratory models that, in case of a direct contact of high-level waste brine, the leached out radioactivity would be disseminated very quickly horizontally and vertically. The reason for this transport mechanism is to be seen in the temperature increase and in the density change of the brine. Up to now no convection streams could be measured directly in the field. (Velocity of flow of the present probe : 1 cm/s).

At present the result of the measurements cannot be interpreted in a satisfying manner. By this reason it is not possible to make predictions about the origin and the behaviour of convection streams in flooded underground workings of a salt mine. Therefore it was decided to postpone the development of numerial methods until more and better understood experimental data are available. Furthermore, a group of the Bundesanstalt für Geowissenschaften und Rohstoffe (BGR) is developing a computer model which should describe the events after an inrush of water into a repository for high-level wastes.

7.3. Disposal of Radioactive Waste in Salt Domes (ECN)

7.3.1. Objectives

To investigate the possibilities of disposal of radioactive waste of all categories in geological rock salt

formations, present in the north-east of the Netherlands. The project will include in the next ten years fieldwork for geologic and geohydrologic examinations, exploratory deep drillings and associated studies, design studies and hazard assessment.

The following work has been done in 1977 :

. calculations concerning heat transfer from the waste to the rock, convergence of deep boreholes, radiation damage effects on salt and thermal migration of small brine inclusions ;

. design studies ;

. studies concerning radionuclide migration and the safety analysis.

7.3.2. Determination of the ability of the formation and of the rock to satisfy the integrity criteria

7.3.2.1. Heat transfer study

Consideration of the possible presence of minerals in the formation other than NaCl together with thermal expansion effects and in the operational field, the working area (mine gallery) temperatures, has led to rather stringent temperature restrictions, with maximum allowable bulk salt temperatures not far above 100°C. Using literature data of thermal properties of NaCl 3-dimensional temperature distributions have been calculated for a range of stacking arrangements and borehole patterns (7). In this way single layer and multi-layer geometrics have been considered as well as waste container stacking heights of 6 - 50 m.

The time dependence of the temperature distributions was also considered for periods from 5 up to 250 years after burial. Initial heat production per waste cannister of 20 cm diameter and 2 m length was taken as 600 W. A small part of the calculations was directed to the temperature distributions arising from disposal in deep boreholes of 600 m length.

7.3.2.2. Convergence of deep boreholes

Preliminary calculations on convergence of deep boreholes have been made.

7.3.2.3. Radiation damage effects on salt

Some work was done to evaluate radiation damage effects on salt and HLW containing glass as well as some other phenomena related to geologic disposal.

7.3.2.4. Thermal migration of small brine inclusions

Interesting results could be deduced from a literature study on thermal migration of small brine inclusions, where it was shown on the basis of interface kinetics and the influence of grain boundaries and dislocations, that in natural salt a limiting temperature gradient exists of an estimated 3-5°C/cm, below which no thermal migration of brine droplets can occur.

7.3.3. Design studies

At the end of the contract period, much attention was given to the design of a repository. This work was done in collaboration with the Department of Mining of the technical University of Delft. The proposal to locate a number of large cavities for bulk disposal of low- and medium-level wastes at or near the working levels of the mine was studied in some detail. Rock salt temperature calculations indicate that an overall initial heat source of 30 MW, quantified on the basis of a total of 1 Million MWe-year nuclear power production, can be accommodated in such a way that, based on the assumptions made with regard to the original rock salt temperatures and the heat source distribution, it will not result in surpassing very conservative design limits of :

60°C in the containment shield of about 200 m rock salt to be maintained undisturbed around the buried wastes,

100°C for the HLW-disposal area globally,

150°C locally directly around the disposal boreholes, and

60°C at the mine gallery levels during the required period of accessibility, not taking into account the cooling capacity of the mine ventilation.

Additional temperature calculations indicate the possibility to simplify the conceptual design by restricting the HLW-disposal to one mining level only, if drilling techniques could be developed for realising dry vertical disposal boreholes from 600 m depth down to about 900 depth.

7.3.4. Waste isolation and repository safety

7.3.4.1. Radionuclides migration

a) Transport model of radionuclides to the biosphere.

Since from the safety analysis it became clear that no quantitative indication could be given on long term hazards in terms of possible dose commitments to future

man under various accident conditions of the repository, a subcontract was given to ITAL to study this aspect. For this a model had to be developed describing the return of radionuclides to the biosphere and to man. The RID was asked to provide a dissolution model for outside water attack, but it soon became apparent that the most pessimistic dissolution times arrived at with the geohydrologic model would be in the range of 2 - 4 x 10^5 years and these time periods, together with the nuclide retention in soils would hardly leave any radionuclide to cause a dose. Therefore it was proposed to consider an additional model wherein flooding of the mine was postulated, a situation that might lead to a faster return of radionuclides. For the biosphere pathway it was easier to make model descriptions and so called "agricultural" and "fishery" models have been developed by ITAL.

b) Measurement of the distribution coefficients of nuclides between water and rock samples surrounding the salt dome (clay)

Also at the start of the programme the effect of soil-retention on return of radionuclides to man was considered to be important. Various measurements of distribution coefficients (K_D) had been published already but the influence of rather strong salt solutions was not known sufficiently. Therefore K_D for soil samples of Dutch origin and mainly samples of clay taken at various depths during drillings in and near salt formations have been measured for Cs, Sr/Y, Pu, Am in 90 % saturated salt solutions. Under these conditions K_D for Pu and Am for clays are still in the range of 5. 10^3 - 9. 10^4.

7.3.4.2. Safety analysis

All imaginable and mostly very unrealistic accident situations have been considered in the safety studies, including meteorite impact, influence of glaciation periods and the like, but the possible routes of water intrusion and its effects were studied most extensively, because these types of accidents were considered to be the most probable routes for breach of containment in the far future. Much effort was devoted to finding a realistic time scale for dissolution of a salt formation in case of contact with water at the flanks and the top. This work led to a theory on self-healing of a formation that was subjected to local attack, by the action of salt-creep that would cause supply of salt to the area under dissolution at a comparative rate, thus assuring a stable geometry.

7.4. Disposal of Radioactive Waste in Clay Formation (CEN-Belgium)

7.4.1. Objectives

Setting up of an experimental storage facility for all categories of radioactive wastes in a clay formation situated under the site of the Centre d'Etude de l'Energie Nucléaire (CEN) in Mol, at an approximate depth of 220 m (Boom clay). Arrangements have been made for an extensive range of in situ confirmation experiments the sinking of an access well and the establishment and subsequent experimental operation of the disposal facility.

The work done in 1977 covered :

- the characterization of site (local geological prospecting and the corresponding rock samples analyses, geotechnical and hydrological prospection ;
- the development of a mathematical model for heat transfer in a clay mass ;
- design studies ;
- studies concerning the waste isolation and the repository safety ;
- the preparation of a file for the authorization application.

7.4.2. Characterization of site

7.4.2.1. Analysis of boom clay samples obtained from the exploratory geological drilling

a) Chemical and mineralogical analysis

The chemical and mineralogical analysis of cores sampled during the exploratory geological drilling carried out in 1975 to a depth of 580 metres were completed in 1977. It is now possible to assess the composition and general properties of the rock across the entire thickness of the formation under consideration between levels of 160 m and 300 m underground.

b) Mechanical and physical properties

The work concerning these properties has also been completed :

- density : average value 1.998 t/m^3, density increasing with depth ;
- grain-size distribution : with one or two exceptions,

all the samples showed grain size less than 200 µ (99 %) ; 55 % of the material was less than 2 µ in diameter ;
- thermal conductivity : the values are in the ranges of 0.30-0.32 W/m°C, at 100°C and 0.58-0.81 W/m°C at 500°C ;
- specific heat : at ambient temperature, the values fluctuate between 0.75 and 0.97 W/g°C and increase to 1.49 at around 300°C ;
- shrinkage during drying : total shrinkage calculated as a percentage of the material in the moist condition varies from 4.7 to 6.5 % ;
- Atterberg limits : on average, 63.86 (limit of liquidity), 23.94 (limit of plasticity) and 39,92 (index of plasticity).

The values for the index for plasticity and the limit of liquidity correspond, according to the Casagrande classification, to a clay of high compressibility with a low content of organic compounds.

c) Other determinations

- Natural radiactovity

The natural radioactivity was measured and the quantity of radionuclides present determined in order to establish the levels of radioactivity and the nature of the radionuclides detected in aggregate during the geophysical measurements. The following radionucldes were determined :

$^{224}_{Ra}$, $^{212}_{Pb}$, $^{208}_{Tl}$ (daughter product of $^{228}_{Th}$) ;

$^{214}_{Pb}$, $^{214}_{Bi}$ (daughter products of $^{226}_{Ra}$) ;

$^{40}_{K}$.

Generally speaking, the content of these various radioactive elements decreases with depth, the clay formations show a greater degree of enrichment than the sandy formations.

- Formation of gaseous products by heating the clay.

The gas analyses were effected either directly in the samples treated specifically for this purpose or during the thermal gravimetry and differential thermal analysis. Interest was focused on the fluorinated and chlorinated compounds and the dioxides of carbon and sulphur. Below 300°C, very small or no releases of fluorinated compounds or carbon dioxide compounds occured ; on the other hand, chlorinated compounds and sulphur dioxide compounds were relased in small amounts.

- Determination of radiolysis products following irradiation of the clay by an intense γ field.

In a first series of experiments, only qualitative determinations of gases formed in the absence of air were carried out. For a γ irradiation dose of 1.10^8 rads, the only element and compound found were hydrogen and carbon dioxide. As these tests were conducted in the absence of air, the phenomena arising corresponded roughly to the following reactions :

$$H_2O \text{ (not fixed or in compound)} \rightleftharpoons H_2 + 1/2\ O_2 \qquad (1)$$

$$CH \text{ organic} + O_2 \rightleftharpoons H_2 + CO_2 \qquad (2)$$

As the oxygen released following reaction (1) does not recur in the gas analysis, the assumption is that it is consumed by reaction (2). In more elaborate tests, quantitative analyses were carried out on the gases released. While the results varied considerably, depending on the techniques used in the treatment of the samples, it was nevertheless clear that, in the case of unheated clay, the gases released consisted mainly of CO_2 and H_2. CH_4 and N_2-Ar were also detected but in small quantities (close to zero). When the clay was heated, the CO_2 content in the gases released was small or non-existent, of the same order of magnitude as the CH_4 and N_2-Ar, the bulk of the gas consisting of hydrogen.

7.4.2.2. Exploratory geotechnical drilling, hydrological prospecting and corresponding characterization

The object of the study was to gain information on the geomechanical properties of undisturbed Boom clay and the properties of the water-bearing layers present in the Mol area.

a) Exploratory drillings and tests on undisturbed samples.

The exploratory drilling was commenced in April 1976 and was completed in October of the same year. Undisturbed samples were taken in the clay at depths between 173 m and 261 m. The length of the samples varied from 20 to 44 cm at a constant diameter of 10 cm. The following tests and determinations were carried out on 50 samples :

- volumetric weight : results between 1.914 and 2.105 (t/m^3)
- dry weitht (t/m^3) : " " 1.484 and 1.728
- water content (%) : " " 17.6 and 28.9
- volume of pores (%) : " " 44 and 34.6
- degree of saturation (%) " " 100 and 88.4
- coefficient of permeability(cm/s) $1.2.10^{-7}$ and $7.1.10^{-10}$

The triaxial cellular tests carried out on all the undisturbed samples gave the following results :

Consolidated undrained test :

- apparent cohesion c' = 1.19 kg/cm^2
- apparent angle of friction φ' = 19°

Non-consolidated undrained test :

- cohesion C_u max = 13 kg/cm^2 and C_u, min = 3.25 kg/cm^2
- angle of friction φ_u = 0.

The Atterberg limits were repeated for a few samples obtained from this drilling. The result of the plasticity index were in the order of 25 % above those given in 1.2.

b) Hydrological prospection

Detailed knowledge of the characteristics of the aquifers present in the potential zone for an underground facility for the storage of radioactive wastes is indispensable to the risk evaluation study. At the start of 1976, several injection wells were drilled on the experimental site chosen and open piezometers were installed at the following levels underground : 570 m (Maastrichtian tufa), 450 m (Landenian sands), 325 m (Ledian sands), 280 m (Rupelia sands), 180 (Antwerp sands - existing well) and at a few metres depth in the phreatic watertable.

In 1977, tests were carried out with the cooperation of other interested bodies on the lowering of the aquifers cojacent with the Boom clay either in situ (at a level between 270 and 300 m underground, Rupelian sands) or in the immediate vicinity (at a level between 75 and 160 m). The following aquifers were revealed :

- phreatic watertable between 0 and 28 m
- semi-artesian aquifer between 28 and 165 m (absolute piezometric level in the order of 22.5 m).

A pumping test on this aquifer made it possible to calculate the following average hydrological characteristics :

permeability 9.8 m/day or $1.1.10^{-4}$ m/s
transmissivity $85.4 . 10^{-4}$ m^2/s

- confined aquifer between 260 m and 300 m (absolute piezometric level in the order of 21,5 m) with the following hydrological characteristics :

permeability $4.2\ 10^{-7}$m/s
transmissivity $0.105\ 10^{-4}$m^2/s
and coefficient of storage : $4.3\ 10^{-4}$

- confined aquifer in the Bruxellian between 310 and 345 m

- artesian aquifer in the Landenian between 445 and 530 m

- confined aquifer in the Maastrichtian below 570 m.

c) Chemical and radiochemical analysis of underground waters.

Water samples are taken each year at about the same time from the water-bearing layers between levels at 0 and 600 m depth. The object of the analysis is to check any variations in chemical and radionuclide composition as a function of time. These indications will serve as a refence of origin in the event of a burial facility being constructed on the site under study.

The following determinations were carried out :

- Ca, Mg, total Fe, K, Cl^-, $SO_4^=$, $CO_3^=$, $PO_4^=$, F^-, $S^=$, NH_4^+, Sr, Zn, Mn, B, Ba ;
- pH, electrical conductivity, redox potential, dissolved oxygen ;
- total U, total Pu, ^{137}Cs, ^{90}Sr, ^{40}K, ^{226}Ra, ^{227}Rn.

7.4.3. Determination of the ability of the formation and of the rock to satisfy the integrity criteria

Location of faults.

In order to confirm the integrity of the site within a zone of 25 km^2, measurements in the field had been envisaged for the purpose of locating faults, bearing in mind the extent of the work planned and after an examination of the practical possibilities of such a measurement programme.

A detailed preliminary study by infra-red visible light photographs of the site taken from aircraft and by satellite was commenced in 1977.

7.4.4. Design studies

All of the experimental information collected at the site selected made it possible to undertake on a realistic basis a study on the setting-up of facilities for the long-term containment of various types of immobilized wastes. The main objectives of the project (known as Hades project), for which a firm of engineering consultants has been commissioned, are as follows :

- evaluation of technical possibilities for the creation of galleries or cavities in the Boom clay at a depth of about 200 m ;
- feasibility study for the long-term containment in the

geological formation under consideration of various types of immobilized radioactive wastes ;
- preliminary conceptual study of the whole project making possible a preliminary assessment of first costs and operating costs.

The expected duration of the study is 6-9 months. It will thus be completed during the summer of 1978.

In parallel, a simple three dimensional model was developed for heat transfer in a clay mass on the basis of one or more isolated heat sources. In terms of some parameters - such as the thermal density of the source, configuration in space, time and thermal conductivity - this model, adjusted as and when experimental results become available should make it possible to define the optimum geometric configuration of a site for the containment of highly active wastes in relation to the total heat load permissible for safety reasons.

7.4.5. Waste isolation and repository safety

7.4.5.1. Corrosion study

On the basis of the results obtained from the various chemical analysis of Boom clay samples obtained from the exploratory geological drilling (see para. 7.4.2.1.) corrosion tests were undertaken in the laboratory on various metals, alloys and oxides. The object of these tests was to study the corrosive effect of the geological medium (natural clay in this case) on materials likely to be used underground.

An example of one of the reconstituted corrosive media is as follows : per litre of water : 0.0049 ml HCl ; 0.015 ml H_2SO_4 ; 0.0004 ml HF ; 8 mg NaCl ; 22 mg Na_2SO_4 ; 0.27 mg CaF_2 ; 0.44 mg KF ; 0.1 mg $MgCl_2$; pH 3.5. Some 20 samples were brought into contact with air saturated with this solution at temperatures in the order of 50°C and in excess of 100°C. At regular intervals, these specimens were weighed and examined metallographically. First results are already available.

7.4.5.2. Determination of ion exchange properties of clay samples

With regard to the determination of ion-exchange properties, radionuclides ^{85}Sr, ^{134}Cs, $^{239-240}Pu$ and $^{152-154}Eu$ were selected for a systematic determination of distribution coefficients in terms of :

- the pH of the solutions ;
- the concentration of ions present in solution ,
- the depth at which the soil samples were taken ;
- the temperature to which the samples were preheated

(max. 500°C) ;
- the γ radiation dose to which the samples were exposed (max. 3.10^{10} rads).

The tests were conducted on a batch basis. While the results for Cs and Sr were in very good agreement, the same could not be said for Pu and Eu (see Fig. 7.4.1.), where various phenomena, such as plymerization and complexing, interfered in the interpretation of the results. It was therefore necessary to undertake research of a more basic nature. The general tendencies may be summarized as follows :

- the total absorption capacity of the clay investigated is in the order of 20 meq. 100 g^{-1} ;
- heat treatment of the clay has a negative effect on its ion exchange capacity ; the distribution coefficients are in some case lowered by a factor of 10 ;
- γ irradiation has an effect similar to that of heat treatment (probably because of the elimination of natural water by radiolys ;
- the effects of a heat treatment and γ irradiation are not cumulated.

During the latter half of 1977, column experiments were commenced ; the first results obtained indicate absorption capacities similar to those obtained in the batch tests.

7.4.5.3. Development of a mathematical model for ion migration in a clay formation

A first simple three-dimensional model to evaluate the extent of the phenomenon of ion migration in a homogeneous formation was developed and applied to the particular site of Mol, taking into account experimental data available, such as hydraulic gradient, permeability of the formation and distribution coefficients of various radionuclides. The first results show that the Mol site has acceptable characteristics from the migration-barrier point of view as far as the main radionuclides, such as plutonium, are concerned.

7.4.5.4. Risk analysis

This problem is being examined in conjunction with the JRC in Ispra. In a first approach, the intention is to apply the "fault tree" technique to the specific case of the Boom clay formation in the Mol area. To this end, data relating to the general geology, hydrogeology, seismicity, tectonics, climatoloty, etc., of North-West Europe have been compiled. A first mathematical assessement is expected before summer 1978.

7.4.6. Preparation of a file for the authorization application

The object of the file is to obtain agreement in principle from the Belgian authorities for the sinking of an access well and an experimental chamber in the Boom clay formation at depths in the order of 220 m at Mol. The file, which will consist of three main parts :

- a summary of the compilation work and the experimental work carried out and the general conclusions to be drawn from them ;
- the results of the feasibility study (Hades project described in Section 7) ;
- a preliminary risk evaluation is in preparation. It should be presented to the competent authorities late 1978.

7.5. Disposal of Radioactive Waste in Clay Formation (CNEN - Italy)

7.5.1. Objectives

To evaluate the possibility of constructing a pilot disposal facility on the clay formation below the CNEN Trisaia site (South Italy).

The work for 1977 included :

- the characterisation of the Trisaia site by determining the geological and geophysical characteristics of the site and analysing clay samples from the Trisaia rock base ;
- the in situ evaluation of the heating of the clay by heat released from the radioactive wastes ;
- preliminary studies on the various ways and means of disposing of radioactive wastes in the formation ;
- a first evaluation of the risks associated with waste disposal in the formation.

7.5.2. Characterisation of site

7.5.2.1. Geological survey of the terrain surrounding the Trisaia centre

A geological survey has been carried out covering a 15 km-diameter around the Trisaia Centre. On the basis of available hydrographic and geomorphological data, the stratigraphy of the region has been established and characterized in similar palaeographic zones and in

clearly defined orogenic phases, according to the following units, from the most ancient to the most recent : Lagonegro unit Cilento unit, Sicilide unit, expostorogenic complex and postorogenic complex.

The various units are linked by orogenic transfer phenomena (sheet tectonic structure) and fold and fault structures connected with orogenic uplifts of subsidences of considerable magnitude. Stress should be laid on the importance of the Metaponto layer, the last in time which is directly relevant to the formations located under the Trisaia Centre.

The structure of the first 2.000 meters from the surface should consist of marine or complex postorogenic terraces followed by slight marly blue clay of the expostorogenic complex, then variogates clays, and finally Tusa tuffites of the Sicilide unit (Metaponte sheet) which are embedded in the blue clay of the piopleistocène Certain faults affect this sheet with considerable amounts of throws, relevant to the marine terraces of the pleistocene.

With regard to the movements of the shoreline, it has been established that vertical displacements of around 100 meters have taken place during the past 10.000 years.

7.5.2.2. Geophysical studies

The results from the seismographic station (three short-period 513 Geotech orthogonal seismometers) installed on the Trisaia site showed that, because of excessive depth noise on the site, it was necessary to move the station about 5 km to the Centre itself.

The data obtained during the seismic reflection-refraction programme are being interpreted.

7.5.2.3. Exploratory drilling at great depth

The technical procedures for the geological exploratory drilling to 750 m by continuous coring have been determined. The diameter of the drill hole (86 mm) should make it possible to obtain undisturbed clay samples.

7.5.2.4. Preliminary analysis of clay samples from the Trisaia rock base

Clay samples obtained from various exploratory drillings in the interior of the Centre have been analysed at depths from 10 m, i.e., immediately below the sand-conglomerate complex, to 90 m. The following determinations have been carried out :

a) chemical analysis : the average composition of the

clay components is stable and is roughly as follows : 50 % (SiO_2), 14 % (Al_2O_3), 1 % (TiO_2), 7 % (CaO), 2 % (MgO), 1,5 % (Na_2O), 2,5 % (K_2O), 5 % (Fe total, 18-19 % (hygroscopic water) ;

b) mineralogical analysis : carried out on the total weight, on the fraction below 2 microns, this analysis revealed the presence of quartz (10 - 15 %), feldspar (5 - 10 %), calcite (10 - 20 %) and amorphous substances. Some samples contain traces of pyrites and dolomite.

The argillaceous minerals (50 - 70 %) consist of illite, smectite, kaolinite, dolomite and mixed-layer minerals ;

c) thermal analysis : heating to 950-1,000°C revealed extensive loss of water at low temperature for all samples subjected to analysis on total weight (Ci-Ci/al thermocouple), the endothermal peaks for the illite and kaolinite being in the region of 580-600°C and the exothermal peaks for kaolinite in the region of 900°C.

7.5.3. Determination of the ability of the formation and of the rock to satisfy the integrity criteria

7.5.3.1. Experimental studies of the effects of heating on the clays

a) In situ tests

As part of the experiment, a 1.2 kW two-metre cylindrical heater will be placed in a 20 cm diameter drillhole, sunk to a depth of 25 m and located close to the future 750 m drillhole ; its function will be to simulate an equivalent volume of high-activity radioactive wastes after ten years cooling. This experiment, which will commence at the beginning of 1978, will use Heating Code 5 for the processing of the temperatures recorded in seven channels (four thermocouples per channel) located on a cycloid around the central drillhole.
The purpose of the experiment is to determine temperatures and thermal gradients created by a particular degree of heating in the formation, to study the chemical, physical and structural changes in the clay and to confirm the theoretical results of the calculation code used.

At the end of 1977, drillholes for the experiment had been completed and the heater was being tested in the laboratory.

b) Laboratory tests

A cylindrical electric heater with power settings of 20, 60 and 100 watts was inserted into a cubic block of

Trisaia clay of 50 cm side, sealed and heat-insulated. The temperatures measured experimentally with the aid of eight thermocouples are of the same order of magnitude (0.5 - 15 % difference) as those calculated with the Heating Code.

7.5.3.2. Mathematical modelling of the effect of heating in clay formations

The first studies to evaluate the dissipation of heat in the formation over time and to determine the critical phenomena triggered by the temperature gradients have been started. The extension of these studies to rectilinear, two-dimensional and three-dimensional sources is also in preparation.

7.5.4. Design studies

A first evaluation of the quantities of high-activity waste to be disposed of was initially undertaken on the basis of the present Italian nuclear strategy. Assuming that a permanent disposal facility will be brought into service in the year 2005, and allowing for the fact that each container (220 1 volume, 30 cm diameter and 300 cm height) will contain 180 1 vitrified high-activity wastes from 2.25 t of reprocessed fuel, the following preliminary values are obtained :

- number of containers to be disposed of in 2005 : 1.250
- total number of containers to be disposed of before 2032 (ten years after the closing-down of the last reactors) : 5.150
- thermal output from the high-activity wastes stored in the formation : 1-10

7.5.5. Waste isolation and repository safety

A mathematical model on the analysis of risks arising from the migration of radionuclides from a disposal facility has been selected. This model, which is based on spherical diffusion in clays of very low permeability, has made it possible to prepare a method of calculation for the high-speed diffusion of aqueous flux (unidirectional model) ; the solutions obtained have been graphically represented by computer.

First adjustment tests on the fault-tree model devised at the European Community Joint Research Centre, Ispra have also been performed.

7.6. Disposal of Radioactive Waste in Cristalline Rocks (CEA-France)

7.6.1. Objectives

Evaluation of the possibilities offered in the Western part of France by cristalline rocks for the disposal of radioactive waste.

The research programme covered by this contract in 1977 is the following :

- characterization of sites including the setting up of a catalogue of suitable granite massifs in France and the prospecting of three massifs called A, B and C, in view of their utilization for active waste repository ;

- study of a model for radionuclides migration in the geological environment with absorbable and not absorbable tracers. Research on artificial and natural barriers in crystalline environments in static and dynamic systems, in both cases laboratory experiments will be supplemented by several measurements in situ.

7.6.2. Characterization of sites

7.6.2.1. Catalogue of granite massifs in France

Eleven granite massifs were catalogued in the Western part of France and three massifs (A, B and C) selected to permit a comparative study of the suitability of granites with different properties for disposal purposes.

7.6.2.2. Prospecting in massif A

The massif occupies an area of 300 km^2 in the Western portion of a huge, complex batholith (750 km^2) where the granite has a markedly intrusive appearance and the break faults are much less obvious than in the other parts of the batholith.

a) Surface geology

In a first phase, the work consisted in petrographic and structural surveys, laboratory analysis of rock samples (essential elements and trace elements), and the making and examining of thin sections. The second phase involved assembling all the data and mapping the whole area (petrographic and structural maps on a scale of 1/25 000), computer-processing the results of chemical analysis, interpreting the thin section findings, entering the structural data on diagrams and interpreting those data.

b) Geophysical survay

- Geophysical survey has been carried out using refraction shooting to identify the mechanical properties of the rock by showing up the weathered zones, cracking and abnormalities and to explore the subsurface topography of the granite in the region adjacent to the country rock. Four formations were identified in the interior of the massif :

  surface formations characterized by little or no consolidation, arenaceous granite with enveloped blocks, fissured granite and, finally, formations giving a wave velocity exceeding 4500-5000 m/s, corresponding to sound granite. Generally speaking this seismic horizon is reached before a depth of 50 m.

- Electrical and electromagnetic prospecting :

  In the centre of the massif, the results confirm the homogeneity of the massif through the first few hundred metres, once the superficial weathered layer has been crossed. In the northern part, the granites become submerged beneath the country rock without any notable faulting or discontinuity being evidenced.

- Gravimetric studies :

  On the basis of the gravimetric study, the results show that the massif A was subdivided into three areas :

  . heavy area, corresponding to the South and West country rock
  . an extremely light and gravimetrically hetereogeneous area
  . a very moderate light and gravimetrically more homogeneous than the previous one area. In this area, one part seems to be the most homogeneous, without heavy anomalies.

After collating the results of the geological and geophysical studies it was concluded that the massif has the shape of an asymmetrical mushroom, the southern and western parts spilling on to the country rock and the northeastern part constituting the rooting area. As regards fracturing, on the southern and eastern edges of the massif there was a good correlation for fractures N 150°E and N 40° E between the gravimetric discontinuities, the fractures deduced from aerial photographs and the faults observed in the field, but this was not true in the case of the mesofractures running in the N 90° direction, appearing on gravimetric map in the south-central part of the massif, which were not evident by cartographic or photogeological surveys. Nonetheless, the least fractured areas identified on the one hand by surface geological data and on the other by geophysical investigations overlap to a certain extent and reveal a favourable area

where a more extensive study could be carried out.

7.6.2.3. Prospecting in massif B

The north-western area of the massif, occupying an area of 400 km^2, was selected for detailed studies, these will be completed during the first half 1978.

a) Surface geology

A little more than half the Massif has been mapped petrographically and structurally on a scale of 1/25 000.

From the petrographic point of view, the Massif consists of two granodiorites, one comprising biotite only, the other biotite and cordierite in broad bands generally lying in an east-west direction. The country rock consists of Briovenian sandstone and greywacke. The superficial formations consist of granitic grit several metres in depth and a discontinuous cover of loessial silt of varying thickness (0-15 m).

Preliminary observations of structural studies indicate that the essential structural features lie in the east-west direction.

b) Geophysical studies

The 1977 programme of geophysical studies in Massif B consisted in assembling all the data that had been obtained hitherto and establishing 1000 new gravimetric stations. As regards magnetism, the only map available for this region is the General Survey of France, drawn on a scale of 1/200 000, from a flight altitude of 3000 m : no anomalies could be identified from a map of this scale. No useful data were obtained from seismic or electrical surveys. Gravimetric data obtained from the 1955 survey using 1.100 measuring stations, were supplemented by data from 1.000 new stations which provided a good density of measuring points. Maps of the residual field and vertical gradient revealed the contours of the massif and enabled the prospected region to be divided into five main areas ; however, a number of anomalies were detected within the massif, the interpretation of which will depend on a subsequent petrophysical study of the various facies of the granite and its country rock.

c) Hydrogeological studies

The programme of hydrogeological studies was begun in September 1977 and comprised the cataloguing of existing water points, the selection and hydrological and hydrogeological study of an experimental catchment basin, and the preparation of a mathematical model of ground water runoff in that basin.

The work undertaken in 1977 is represented in Fig. 7.6.1. Approximately 150 wells were catalogued ; they were generally fairly shallow (8-9 m) showing that the weathered part of the granite is not very thick. Within the Massif a basin clearly delimited by rivers (Fig. 2), with an area of approximately 25 km^2, was selected. A permanent station at the basin exit measures the flow continuously ; during the low water period, the distribution of flow throughout the basin was evaluated by two measuring campaigns. A programme of exploratory drilling to a depth of 75 m is now under way : the results will permit a comparative study of the piezometric levels of the water table in the weathered part and those of the table in the fissured sound granite. Finally, a hydrogeological study has been carried out from the test station in order to study transfer mechanisms in Massif B (Fig. 7.6.2.)

7.6.2.4. Study of Massif C

The geographical location of Massif C2 merits special attention. The granite forms part of a complex region made up of a pre-Cambrian crystallophyllitic basement and a shaly sandstone cover of Cambro-Ordovician age. The rocks in the basement are heterogeneous and much disrupted by tectonic activity ; contact between the basement and the sedimentary overlay is generally complicated by thrust faults and tear faults. The field survey amplified and confirmed the available data, and provided a basis for the planning of the study.

7.6.3. Waste isolation and repository safety

7.6.3.1. Study of transfer mechanisms of radioactive products in the environment

Model of migration of radioelements in the ground. The work of modelling consisted mainly in developing the three-dimensional migration model based on isoparametric finite elements. The model consists of one module for calculating velocities and hydraulic pressures in the medium, and one module for calculating migration, taking into account convection, dispersion, linear and reversible absorption and radioactive decay.

With its finite-element parallelipiped linkage it is capable of representing any geometry in three-dimensional space. When the validity of the model was tested for various flow-zone geometries, difficulties were encountered in calculating the convective flow velocities, which induced us to reformulate the definition of the velocity field within the medium : the flow calculation module, which provides the value of the pressures and, subsequently, their derived values at the nodes of the linkage, is now combined with a module for calculating

velocities within each element, by a function of polynomial approximation of the third order (analoguous to the function of pressure approximation fitted to the nodal velocity values). This velocity field, which is now continuous in each element and from one element to the next, is then introduced into the module of convective displacement. The results of the calculations of migration thus obtained are considerably better.

The representation of absorption in cracked media was introduced by using a coefficient of distribution through fracture, expressed as a quantity absorbed per unit of gross fracture area (ml/m^2) instead of the usual unit of mass of porous medium (ml/g). This coefficient was measured in a core sample taken from the selected experimental area, by the BRGM.. The density of fracture of the medium must also be obtained.

A start has been made on interpreting the results of the first set of field tests ; this work is still in hand and will continue, to include simultaneous interpretation of the second (November 1977) set when the results are available.

Finally, the one-dimension migration model has been modified to take account of the simultaneous displacement of harmful elements generated as a result of daughter-product formation during radioactive decay of the migrating elements (e.g. radium or uranium derived from plutonium, curium and americium).

- Experimental testing on Massif A

a) Experimental conditions.

The test station fitted out in 1976 consists of a group of boreholes, drilled to an approximate depth of 40 m, laid out as in Fig. 7.6.3. The central borehole contains a submerged pump capable of inducing convergent radial flow of the groundwater. In the four cardinal directions, satellite boreholes are drilled at approximately 12 m from the central point. None of the boreholes is cased. One has been cored so that the state of fissuring can be assessed : after a weathered layer of some 10 m, a layer of horizontal fissures extends to a depth of 20 m. However, it appears that vertical fissures may extend throughout the explored depth.

b) Experiments and results

The tests (two two-week periods) consisted in constant-rate pumping in the central borehole and short-duration injections into each of the satellite wells. In the first series of tests, the transfer of substances regarded as good water tracers (iodide, chloride, nitrate, thodamine, aramine) were studied ; in the second series, the behaviour of soluble substances close to the products to be stored (caesium and strontium salts) was studied.

On the basis of these tests, the characteristics of the medium were identified.

- Hydrodynamic properties.
  The tests show that the hydrodynamic parameters are characterized by very marked anisotropy apparently resulting from vertical East-West fissuring.

- Transfer patterns.

By means of a radial transfer model, kinematic porosity was evaluated and the equivalent coefficient of dispersion for the various substances studied especially strontium and calcium which may be present in the products for storage.

On the basis of the results, the following main conclusions may be drawn :

. The behaviour of the experimental fissured Massif may be represented as in Fig. 4 : the superficial part, where fissuring is extensive and relatively isotropic (the Southern area being extremely clayey) and the solid deep-seated part with a vertical East-West fissuration providing a direct link between P4 and P1. Thus, the hydrodynamic reactions in the superficial part, for piezometers P2bis, P3 and SR, are slight and fairly independent of the flow, although the transfer or products does take place (except for P3). Between P4 and P1, very direct hydrodynamic links and rapid migration of the substances. This schematic representation will be used for mathematical modelling.

. Substance transfer characteristics.
  As regards the transfer properties of the substances, the plotted curves representing the migration of the products have not all been fully interpreted ; but it is possible to draw the main conclusions as regards the influence of the route and the influence of the nature of the product.

α) Influence of the route : in this complex system migration obviously depends on the point of injection. The interpretation of the radial flow signals gives the following values :

Route	Tracer	U (m/h)	α (m)
P3 - P1	Rhodamine WT	0	X
SR - P1	I^-	0.062	0.8
P2bis - P1	NO^-	0.10	3.5
P4 - P1	Cl^3	9.0	1.5

where U is the apparent mean velocity of the vector fluid between the points of injection and recovery, and α is the coefficient of dispersivity (assuming longitutinal dispersion coefficient $D = \alpha U$). These observed values confirm the very heterogeneous character of the massif as a whole.

β) Influence of the nature of the product : generally speaking, effects of exchange with the weathered materials coating even the deep-seated fissures were observed.

The effects are less marked on anions than on cations. They cause a disappearance of the injected product at the end of route P4-P1 (12 metres) only 31 % of the strontium and 29 % of the caesium were found, compared with 72 % of the iodide and 75 % of the chloride.

γ) Tests on rock samples

Migration tests in several fissured granite samples, obtained from a depth of 29 m in the cored borehole, were conducted in the laboratory. The experimental conditions were controlled to reproduce the order of magnitude of the velocities and concentrations taken into account for the same substances in the tests in situ i.e. ; nitrate chloride, iodide, uranine (fluoresceine) and rhodamine NT as tracers and strontium (chloride) and caesium (sulphate) as telltate storage subsances. The tests confirm and throw light on the observations made in the field :

- the mediocre properties of the classic tracers used in hydrology, such as xanthenic dyes, are confirmed. These tracers should not be used in future experiments ;

- the products are differentiated mainly on the basis of their ionic charge : the cations, contrary to the anions, are subject to delay and ablation. This occurs characteristically by absorption on the clays coating the fissures.

X-ray analyses of the whethered materials show the presence of montmorillonite, plagioclase, quartz, muscovite and goethite. Furthermore, by studying the fracture surfaces with the aid of a quantitative image dissector, it was possible to evaluate the proportion of the surface coated with weathered materials, which is of the order of 16 % after the leaching involved in the various operations to which the sample was subjected.

7.6.3.2. Study of artificial and natural geochemical barriers

For the purposes of underground storage in a crystalline medium, it is necessary to study the geochemical

phenomena (water-rock interaction) that occur after the leaching of granite massifs by slightly mineralized water in order to select the most suitable materials for constituting an artificial geochemical barrier to understand the physico chemical processes of interaction between long-lived radioactive elements and artificial or natural geochemical barriers and to predict by computer simulation the geochemical behaviour of these elements as they travel across the various barriers.

a) Properties of the materials studied.
Twelve absorbent materials were selected and the following properties determined : chemical composition, particle size, exchange potential, mineralogical composition.
The results of the chemical analyses are shown on Fig. 5
The exchange capacities are as follows (in $Ca2^+$ meg/100 g) :

Samples	Exchange capacities $Ca2^+$ meg/100 g
Magadiite	68,31
Attapulgite	36,82
Illite	23,63
Silice	3,78
Kaolinite	3,79
Clinoptilolite	24,06
Bentonite	64,26
Sepiolite	N.D.
Vermiculite	= 100

The mineralogical composition was determined from the chemical composition and from differential thermal analyses, thermo gravimetric analysis and X-ray diffraction. These analyses were conducted only on natural aluminium silicates.

b) Experimental results in static conditions.
By testing the sorption, in static conditions, of the various radioactive ions on highly-sorbent minerals one can determine the distribution coefficients K_d of substrate-solution pairs as a function of pH, and then construct the fixation isotherms. The results obtained at the buffer pH value for each pair give q qualitative indication of the sorptive power of the materials with regard to the elements considered (Fig. 6). The distribution coefficients were classified per pH unit (5 to 9).

In the light of these various results it appears that further studies of artificial barriers should concentrate on the following materials : attapulgite, bentonite, clinoptilolite, illite, kaolinite and vermiculite.

c) Experimental results in dynamic conditions

Sorption tests in dynamic conditions allow study of the

interaction between substrate and solution during flow. These experiments are conducted with the aid of columns, measuring 20 cm in length and 4 cm in diameter, in which flow takes place at a rate of 30 ml/h. So far the pairs caesium-quartz and caesium-quartz with vermicullite have been studied. It is seen that, for the same flow, the behaviour of caesium is very different in the presence of vermiculite ; in the case caesium shows considerable delay in reaching the exit of the column and the concentration is attenuated. This shown the importance of introducing a 1 % vermiculite barrier into the siliceous substrate.

7.7. Disposal of Radioactive Waste in Cristalline Rocks (UKAE -UK)

7.7.1. Objectives

To study the possibility of safe and ultimate disposal of highly radioactive and/or alpha bearing vitrified waste in Great Britain. The study should provide sufficient information to allow initial assessment of possible disposal sites and to identify at least one such site for detailed further investigation.

In 1977, the programme has been concentrated on ;

- geological and associated surveys for selection of possible site
- initiation of associated works, such as petrological, rock mechanics, hydrological and thermal studies
- supporting engineering research to investigate the significance of non-geological factors in the overall feasibility of the proposed disposal procedure
- waste isolation and possible migration of radionuclides through the rocks and to the biosphere.

7.7.2. Characterisation of sites

7.7.2.1. Site selection

The UKAEA has sub-contracted the Institute of Geological Sciences to advise on areas within the United Kingdom which are potentially suitable for research studies and to institute field work on selected sites. 13 areas were selected for review on the basis of a desk study by IGS. These areas were initially selected partly on their geological characteristics but also taking account of non-geological factors such as land ownership. Thus the overall 'suitability' of the areas differs from that in the catalogue prepared for the EEC under a separate contract, where non-geological factors were

excluded from consideration. Commencing in mid-1976, detailed desk studies of the selected areas accompanied by field reconnaissance visits were initiated to collect available geological and physical information relevant to site evaluation, in preparation for field work in 1977 and as a means of selecting the most promising areas for full scale site evaluations. Each area has been reviewed as to petrographical features, structural settings and proximities to mineral deposits, quarries and hydro-electric or water supply schemes. The areas selected fall into three categoreies, based on a combination of technical and political factors :

- areas which appear to have the most promising characteristics and which invite early site investigations ;

- areas which require follow-up investigations in the field to confirm their suitability ;

- areas which have known adverse geological characteristics which makes them unlikely to be suitable although they should not be finally rejected without a detailed area reconnaissance.

It is desirable that geological studies should proceed simultaneously on at least 3 sites of differing geology as follows :

- a Caledonian granite, as the Loch Doon Granite (7.7.1)

- an area of Liwisian basement rocks, and

- an area of migmatized Moine rocks.

Planning consent is currently being sought for borehole drilling to start on at least two sites in early 1978 since much of the necessary study at a desk level has now been completed, including geological interpretation of aerial photography.

7.7.2.2. Petrological and geochemical properties

A detailed field investigation programme has been drawn up to define the best approach to a detailed petrological and geochemical study of any proposed research site. This programme includes development of sub-surface and three-dimensional mapping techniques from surface and borehole date, geochemical characterisation by a detailed analytical programme, laboratory investigations and interaction with research.

As part of a preliminary investigation of the Loch Doon comple , a detailed literature and rock specimens search has been made to locate all available data on the area.

7.7.3. Determination of the ability of the formation and of the rock to satisfy the integrity criteria

7.7.3.1. Petrological and rock mechanics studies

The petrological study has largely comprised theoretical appraisals of the effects of a proposed repository on various crystalline rocks and an experimental programme has been proposed to examine the effects of warm fluids on the host rock under the pressure/temperature conditions of a disposal site. This study will take into account fluid/grout interactions and be run in conjunction with a rock mechanics experimental programme to study changing rock properties with fluid reaction (Fig. 7.7.2.).

7.7.3.2. Hydrogeological studies

a) Hydraulic properties of poorly permeable fractured media.
If the migration of radionuclides as a result of rock melting is discounted, groundwater is the most important agent capable of transporting waste material away from a burial site in significant quantities. The emergence of contaminated groundwater into man's environment will be a function of the magnitude and density of discontinuities within the crystalline rocks. Flow within these discontinuities is likely to be considerably modified by the construction of a disposal facility but information on the natural conditions will enable predictions to be made of likely fluid movement. This it is important that the rocks are characterized in terms of their capacity to transmit and store water. The techniques available for the determination of the hydraulic properties of poorly permeable fractured media have been reviewed and suitable equiment investigated (Fig. 7.7.3.).

b) Water dating methods.
Water dating methods are being investigated, utilising measurement of uranium and uranium-thorium isotope ratios since they can indicate the residence time of formation water. Long residence times are usually associated with zones of low groundwater velocity.

The sensivity of the dating method has been estimated and it has been established that the lower limit of the dating method depends on the uranium content as well as on the lambda value of the particular isotope. A lower limit of 540 years has been obtained for the 230Th/234 U method for a 100 µg uranium sample.

7.7.3.3. Thermal studies

To reduce leaching from vitrified waste, corrosion of canning materials and the migration of radioactivity

through the rock, it is necessary to limit the temperature rise due to fission product heating in waste. At present, geological and chemical evidence suggest that bulk rock structures should not be allowed significantly to exceed 100°C and modelling studies have been conducted to establish the sizes and shapes of block distributions which will allow such limits to be met. Preliminary calculations indicate that the waste which will have accumulated by 2000 AD from the existing nuclear power stations will need to be dispersed through about 1 cubic kilometre of rock. To ensure that the volume required for a repository is kept to a minimum there is a need to develop more reliable data on heat transfer throughout and around reposotories and this work is now in hand.

a) Field experiment.

An experiment has been established in Cornwall where the techniques and instrumentation necessary to measure thermal diffusivity, thermal conductivity and rock mechanical properties can be developed. Since rock structures concerned would be unsuitable for the construction of a repository, it is intended that instrumentation developed at this site will be later transferred to one of the geological research sites to be identified by the Institute of Geological Sciences. An 18 kw electrical heater capable of simulating vitrified waste blocks has been constructed and is being installed in an existing borehole on the research site. It is surrounded by a series of thermocouple holes to enable monitoring of rock temperature changes.

Initial results have been obtained during a five day period of operation (11 Kw power) at a 10 m depth, the rise in temperature of the edge of the heater hole and in hole from 2,5 to 10 ft distance radially from the heater hole on plotted against them as pointed in Fig. 4. Theoretically predicted temperatures calculated assuming a constant laboratory measured value of 3,3 W/m°C of the conductivity for the Carnmenellis granite are shown as lines : general agreement between the experimental and theoretical values can be seen.

b) Assessment of the thermal effects in the rocks. There are several such effects including :

- thermal stressing and cracking which may increase the rock permeability and waterflow
- natural conversion of water in the pores and fissures of the rock
- changes with temperature in adsorption of radioactivity and migration through the rock.

Initial calculations have been completed on the rock mechanical properties of a hard rock repository. The analysis indicates that resultant tensile stresses in

the rock overlying a repository may be sufficient to cause fracture of the rock possibly leading to leakage of radioactivity. In addition, compressive stress through the repository may cause collapse of tunnels in the rock more readily than would normally be the case in mines without heating. Such effects will be sensitive to the actual temperature rise within the repository which will probably reach a maximum value about one hundred years after deposition. It can be reduced by burying the waste at a greater depth and by reducing the bulk average temperature in the rock.

The work is being carried out in close collaboration with related experiments in Sweden, France, Belgium and Germany.

7.7.4. Design studies

A conceptual design study for an underground repository is being prepared, assuming that retrieval of disposal material will not be required and that the repository will be at least 300 metres below the surface.

7.7.4.1. Evaluation of the waste to dispose

Consideration is being given to the dimensions of the individual waste blocks requiring disposal and to their thermal and radioactive properties. The present dimensions proposed for the canisters (0.5 metres diameter by 3 metres long) combined with the high heat release from fission product decay (the heat output per canister at vitrification is limited to 11 kW), leads to major problems in designing a repository. Handling such large containers underground, particularly when in association with shielding, will lead to a highly capital intensive facility. In addition long periods of storage will be necessary to enable sufficient fission product decay to take place to enable the temperature constraints on repository structures to be met. The present indication is that vitrified wastes will require approximately 50 years storage, prior to disposal. The total heat output from an individual block will need to be reduced to about 1 kW before disposal will be practicable, when the blocks could be emplaced in a granite formation in a cubic array with a 20 metres spacing between adjacent blocks. In this study it is assumed that the waste arising from the UK nuclear power programme by the end of the century will be equivalent to between 2000 and 3000 glass blocks in form of harwest type canisters.

7.7.4.2. Design Studies

- Emplacement holes.

A preliminary study of excavation methods suggests that

emplacement in the cubic array could be achieved at minimum cost by driving 5 m diameter horizontal tunnels at depth with vertical holes from the floor about 150 m deep, each hole containing a number of glass blocks. This will minimise the length of horizontal tunnels which are likely to represent the major excavation costs. Vertical emplacement holes should be drilled rather than blasted to prevent undue stressing of the surrounding rock structures. A survey of available rigs has indicated that equipment is available for drilling 0.5 m holes in hard rock at a cost of approximately £50 per metre. The equipment would need extensive modification for use underground in galleries with limited headroom.

Consideration may need to be given to the possibility of surrounding individual canisters with a heat transfer or barrier medium to control migration of heat or nuclides into surrounding structures. Construction of suitable holes of 1-2 metres diameter could best be achieved by the raise boring technique which requires two horizontal galleries at the top and bottom of the disposal shaft. This technique is now well estalbished but is several times more expensive than direct drilling and leads to a large increase in the spoil production.

Access shafts

Access shafts for men, materials and blocks could be constructed using existing techniques down to a depth of 2 kilometres. Current costs for such shafts is likely to be between £3000 and £6000 per metre. There may well be advantages in creating inclined or horizontal access shafts if the local topography proves to be suitable for their use (Fig. 7.7.5. and 7.7.6.). Present plans are to place as many of the service facilities as possible underground to minimise environmental impact on the surface. It is anticipated that the detailed design may restrict visual intrusion to the access shaft and 2 or 3 ventilation shafts.

Disposal of spoil.

To minimise the need to dispose of spoil on the surface and to provide continuity of employment it is attractive to consider if repository construction can continue whilst deposition is taking place in parts constructed in earlier phases. Backfilling can then be used to close off shafts in parts of the repository where the waste has already been isolated. Galleries and other workings may well prove to be suitable for disposal of medium and low level wastes with the spoil.

7.7.5. Waste isolation and repository safety

7.7.5.1. Material corrosion

Analytical methods suitable for measuring chloride in

granites have been established : the samples measured contained chloride in the range 130 to 225 µg/g.

Experiments are in progress to investigate the corrosion of various materials (Fe, Ni, Ci austenitic alloys) which may be used to sheath the vitrified waste in an underground repository. For this purpose it has been necessary to produce a liquor resembling the groundwater which would be present within the pores and interstices of granite structures. This has been achieved by extracting soluble salts from powdered granite with distilled water at temperatures up to 120° C. Analysis of the resulting liquor indicates the presence of up to 50 ppm chloride and significant quantities of Na, Sr and So. Rock samples heated to high temperatures (300 to 900°C) only lose chloride in the presence of steam. The granites examined have shown no evidence of unexpected phase changes or other related thermal effects up to 1000° C. It is recognised that natural groundwater may contain much higher concentrations, derived from this and other sources.

Field experiments are also planned in association with the heating experiments in Cornwall, primarily to develop equipment and techniques, in which corrosion specimens will be exposed both below the heater and in a separate borehole. Materials to be tested include the recommended Harvest cladding, steel, lead, copper, gold and titanium. The experimentation will permit temperature monitoring and atmosphere sampling above and around the specimens. It is recognised that the results may not be relevant to an actual disposal site.

7.5.2. Dissolution and movement of radionuclide through the rock

A programme of research into the chemical effects which occur within a repository is in progress. Factors affecting the dissolution and movement of radionuclides through the rock are being investigated using leachate from simulated Harvest glass containing plutonium and americium as well as specially prepared actinide containing solutions. Retention of the nuclides by powdered granite has been investigated. In experiments in which 5 grams of granite was contacted with 25 millilitres of solutions containing 1.75 micrograms of plutonium as plutonium nitrate, or containing 2.5 micrograms of plutonium in the form of leachate from plutonium-containing glass, almost all of the plutonium was removed from solution in 15 days. Experiments using solutions containing 0.004 micrograms of americium gave similar results. Studies are continuing using these solutions to other concentrations, at elevated temperatures and in the presence of substances which may be contained in underground water sources, including silica, chloride and fluoride. Experiments are also being conducted using columns of powdered granite which are indicating similar retention factors. Subsequent release of retained nuclides will be investigated in parallel with the retention studies.

7.7.5.3. Assessment of release of activity and pathway back to man

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The UKAEA has sub-contracted the National Radiological Protection Board to produce a release model and a transport model to assist in assessing the safety of underground disposal. Preliminary results for a range of nuclides which will be present in the waste, making conservative estimates of rates of water movement and delay by sorption phenomena, indicate maximum doses to man which are a small proportion of those currently accepted by the ICRP.
The calculations show that four radionuclides, ^{99}Tc, ^{129}I, ^{226}Ra, and ^{237}Np, give rise to the highest individual and collective doses by the major exposure pathway without consideration of fresh water, terrestral or marine pathways, of direct ingestion of drinking water. The peak individual doses from these radionuclides range from 2 % to 8 % of the dose limit for the relevant critical organ (figure 7.7.7.);
peak annual collective doses are estimated to be of the order of 10^4 man rem to the critical organ and 40 - 500 man rem to the total body. Radionuclides of secondary importance in terms of dose include the long-lived fission products, ^{79}Se, ^{93}Zr, ^{126}Sn and ^{135}Cs, and lower members of the actinide chains such as ^{210}Pb, ^{210}Po, ^{225}Ra, ^{229}Th and ^{233}Pa. Peak individual doses from these nuclides are in the range, 0.001-0.1%, of the appropriate dose limit; maximum annual collective doses are 1 - 200 man rem to the critical organ and 3 - 4 man rem to the total body.

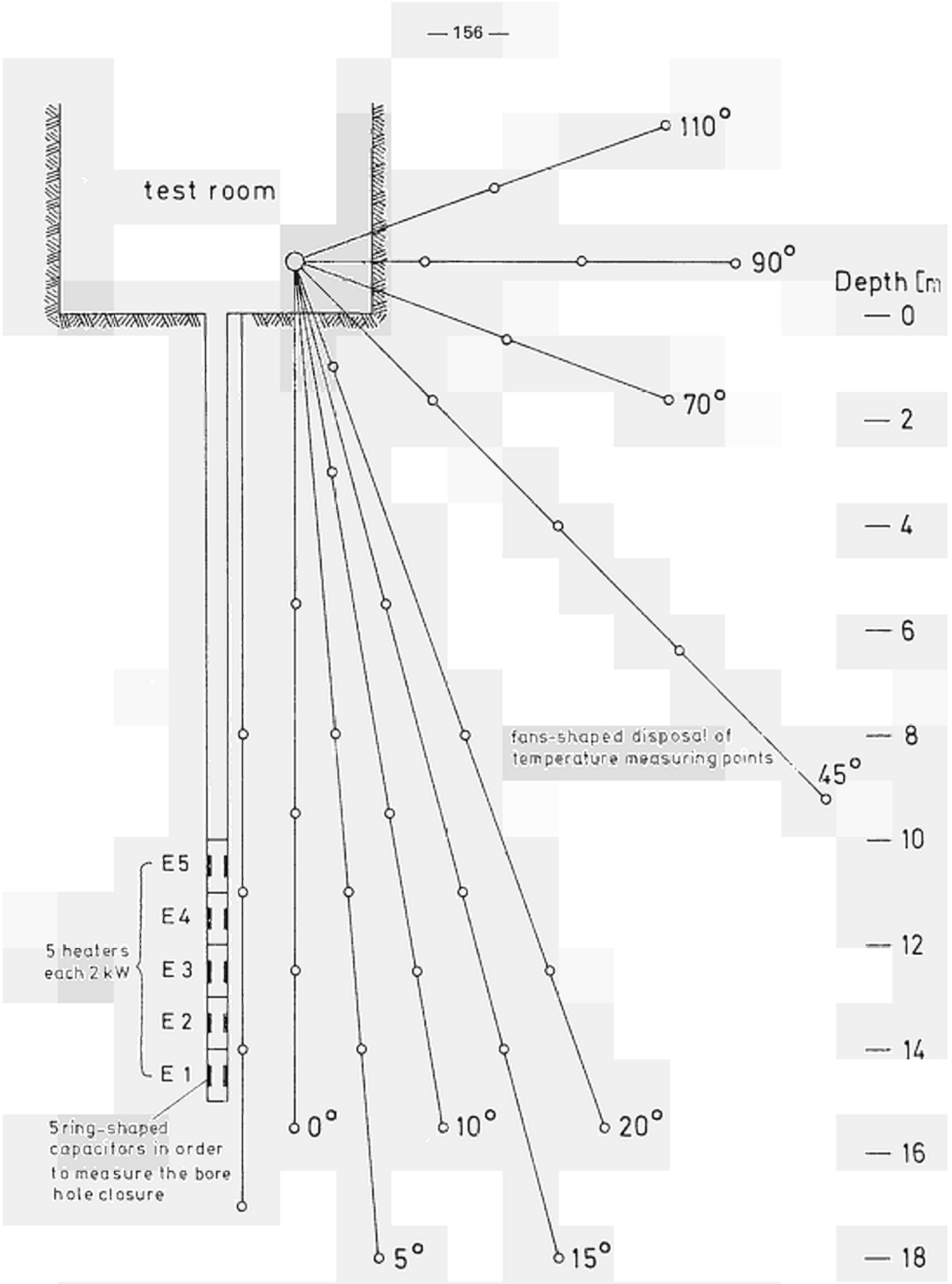


FIGURE 7.2.1 : Temperature test field 4, arrangement of heaters and thermocouples

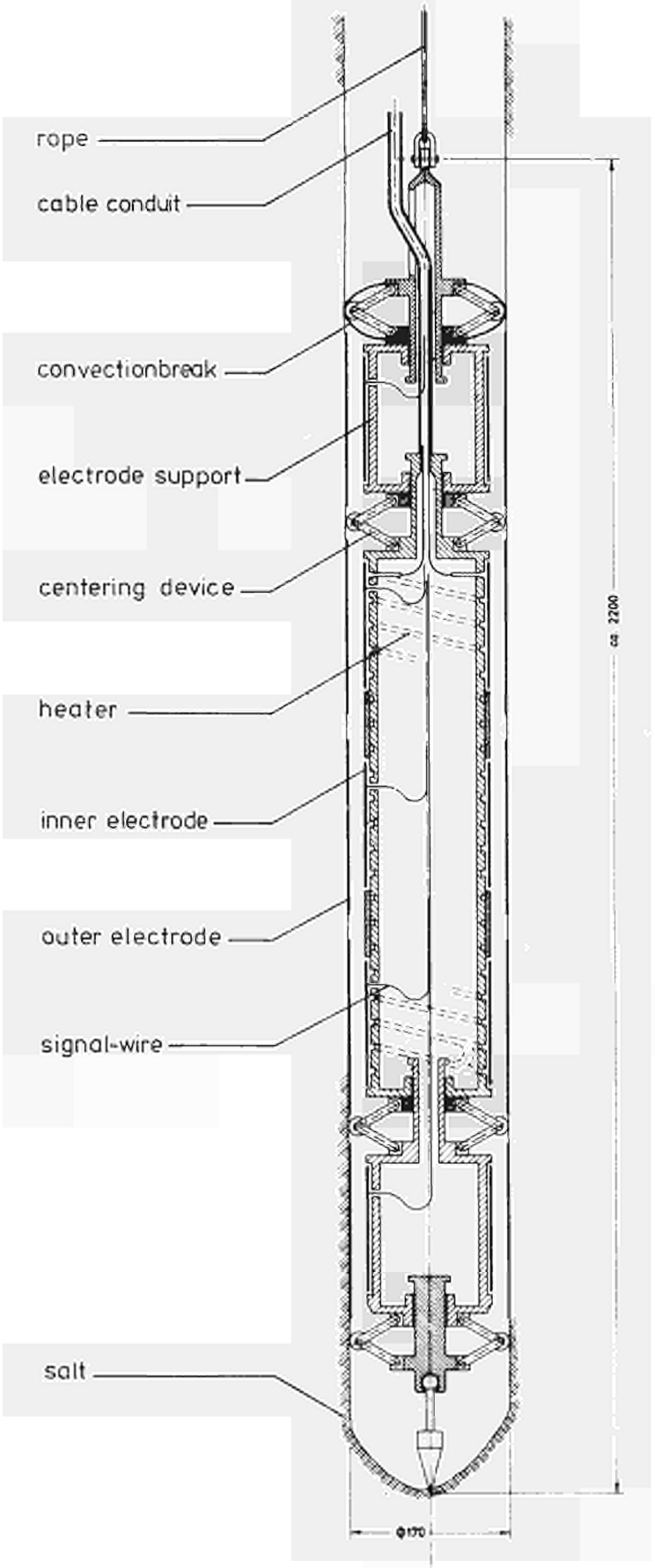


FIG. 7.2.2 : MECHANICAL PRINCIPLE OF THE STANDARD-PROBE

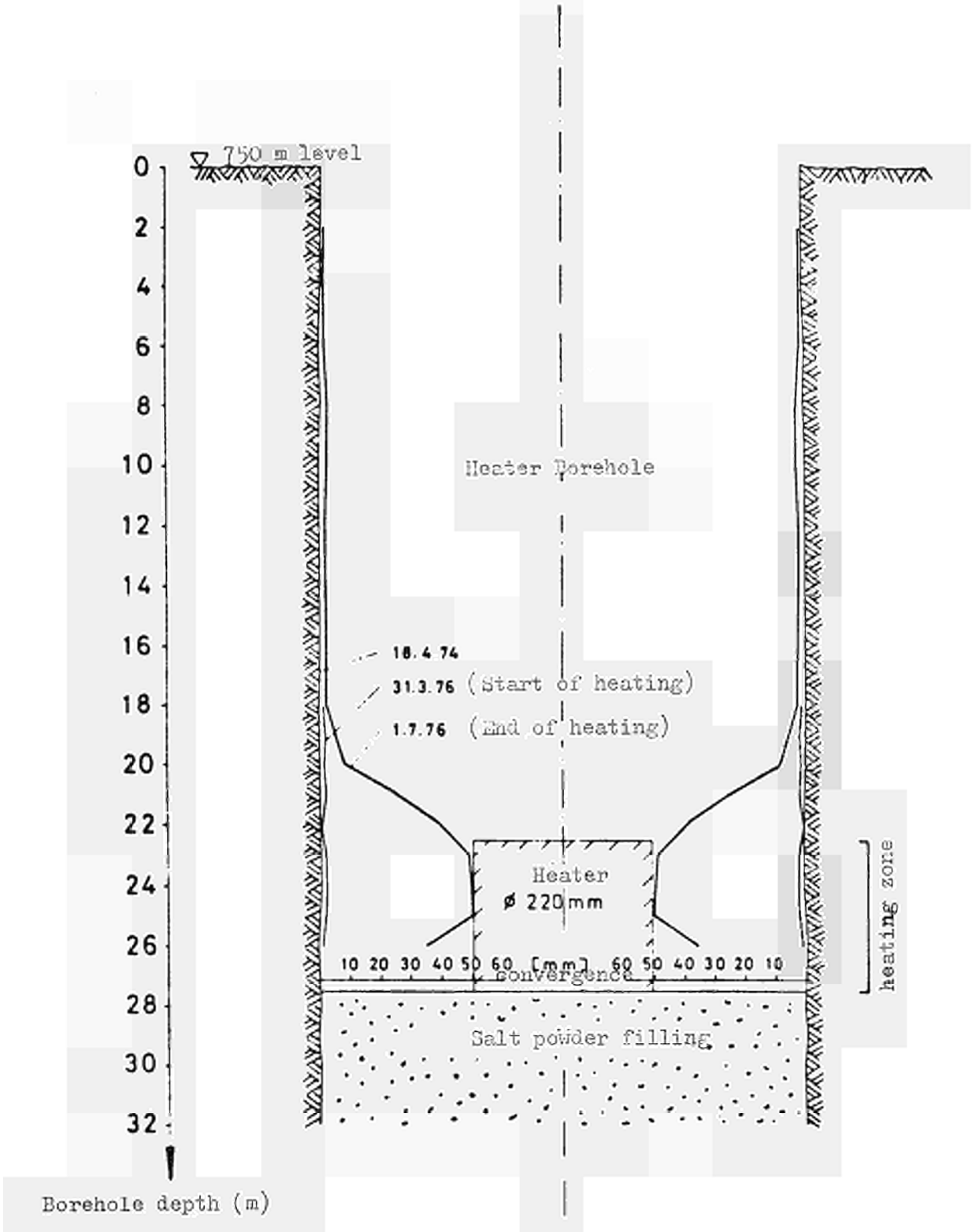


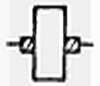
FIGURE 7.2.3 : CONVERGENCE OF HEATER BOREHOLE IN THE ASSE SALT MINE (Temperature-Experiment field)

Fig.: 7.2.4 Material Properties of Asse-Rock-Salt (Series I), determined with the Pulse Transmission Technique

Origin: Asse Mine of GSF, Remlingen/Wolfenbüttel — Salt section: Older Halite (Na 2 Ü), medium grained

Site of sampling: 750 m-level, 2. western cross-cut — Horizontal drill holes in the direction of layers strike

Specimen size: Cylinder, 100 mm dia., 250 mm long, — 15 samples

direction of pulse transmission	material properties / statistical data	density ϱ [kg/m^3]	ultrasonic velocities V_p [m/s]	V_s [m/s]	dynamic elastic constants σ [-]	E [10^{10} N/m^2]	K [10^{10} N/m^2]	μ [10^{10} N/m^2]	λ [10^{10} N/m^2]
 ∥ to the axis of test sample	number of data points n	15	15	15	15	15	15	15	15
	mean $\bar{x}$	2204	4540	2550	0,271	3,63	2,65	1,43	1,69
	standard deviation s_x	± 26	± 62	± 43	±0,011	± 0,1	± 0,12	± 0,04	±0,13
	standard deviation from the mean $s_{\bar{x}}$	± 7	± 16	± 11	±0,003	± 0,02	± 0,03	± 0,01	±0,03
	coeff. of variation V[%]	± 1,2	± 1,4	± 1,7	± 4,1	± 2,6	± 4,7	± 3,1	± 7,6
 ⊥ to the axis of test sample	number of data points n	15	15	15	15	15	15	15	15
	mean $\bar{x}$	2204	4460	2540	0,259	3,59	2,49	1,43	1,54
	standard deviation s_x	± 26	± 111	± 53	± 0,018	± 0,14	± 0,21	± 0,06	± 0,21
	standard deviation from the mean $s_{\bar{x}}$	± 7	± 29	± 14	± 0,004	± 0,04	± 0,05	±0,02	± 0,05
	coeff. of variation V[%]	± 1,2	± 2,5	± 2,1	± 6,6	± 3,9	± 8,4	±4,2	±13,8

V_p = longitudinal velocity (infinite medium)

V_s = shear velocity

σ = Poisson's ratio

E = Young's modulus

K = bulk modulus

μ = modulus of rigidity

λ = Lamé's constant

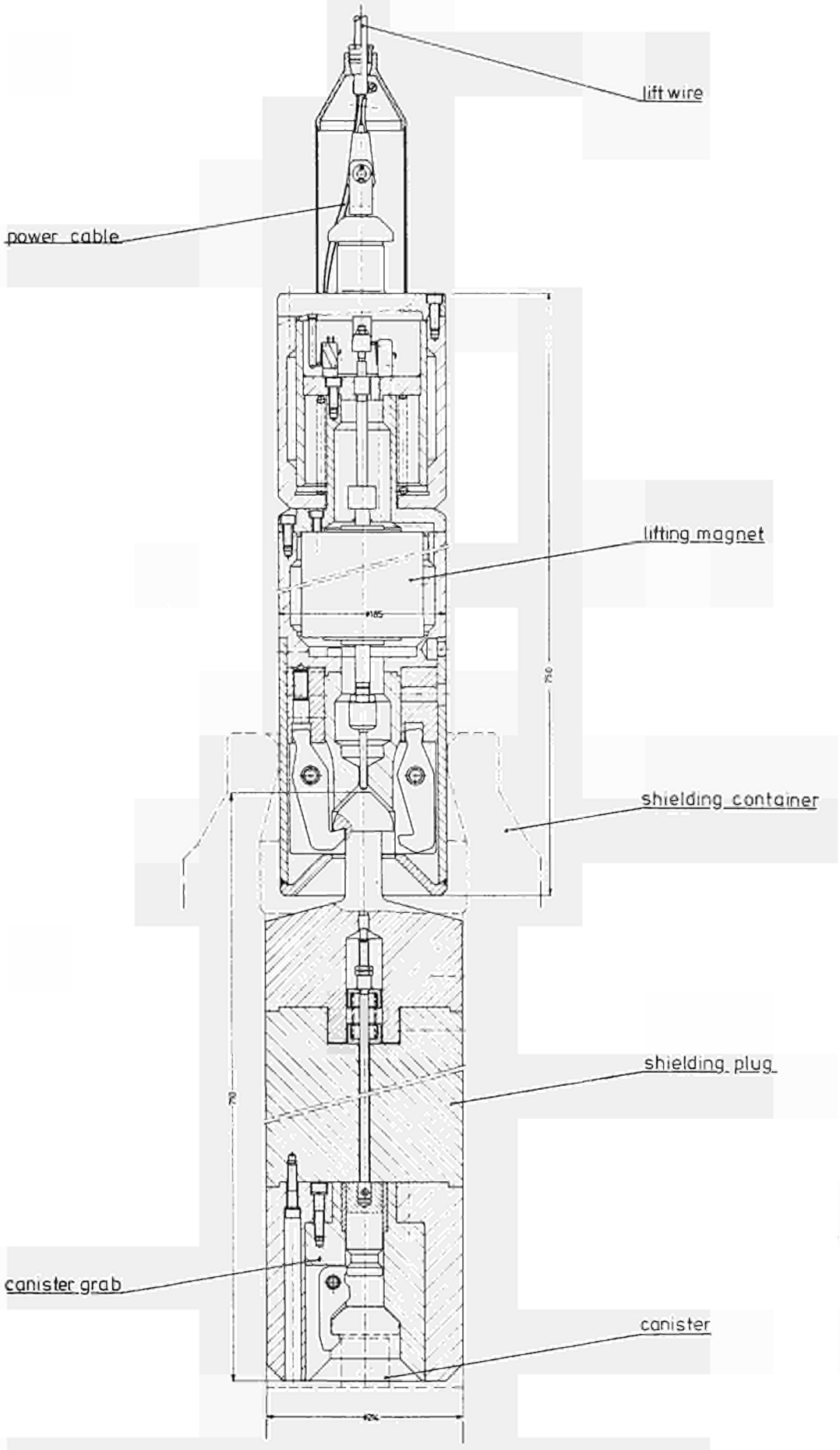
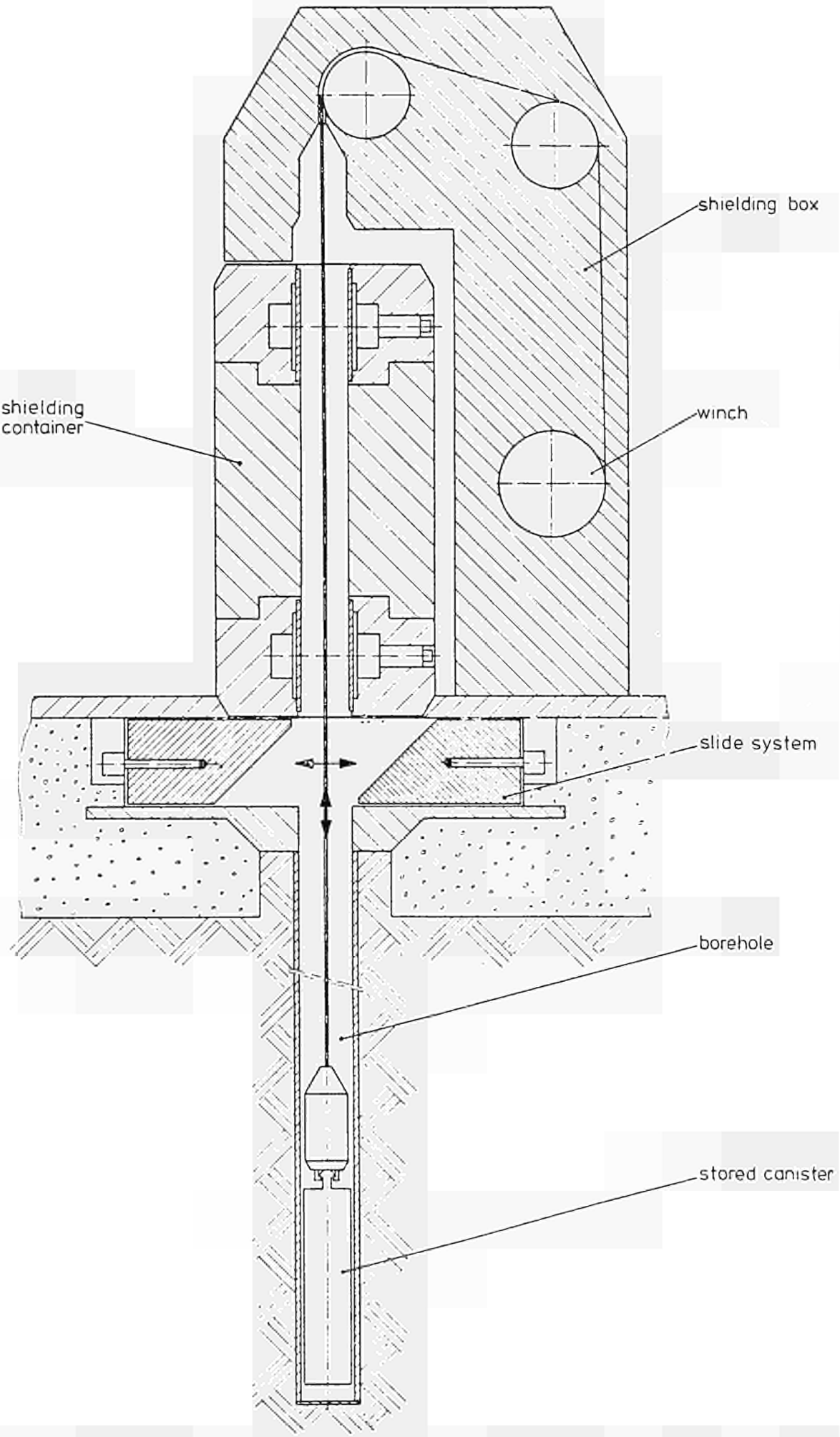


Fig. 7.2.5 : Canister grab system with shielding plug



g. 7.2.6 : Shielding box for unloading the high level waste container

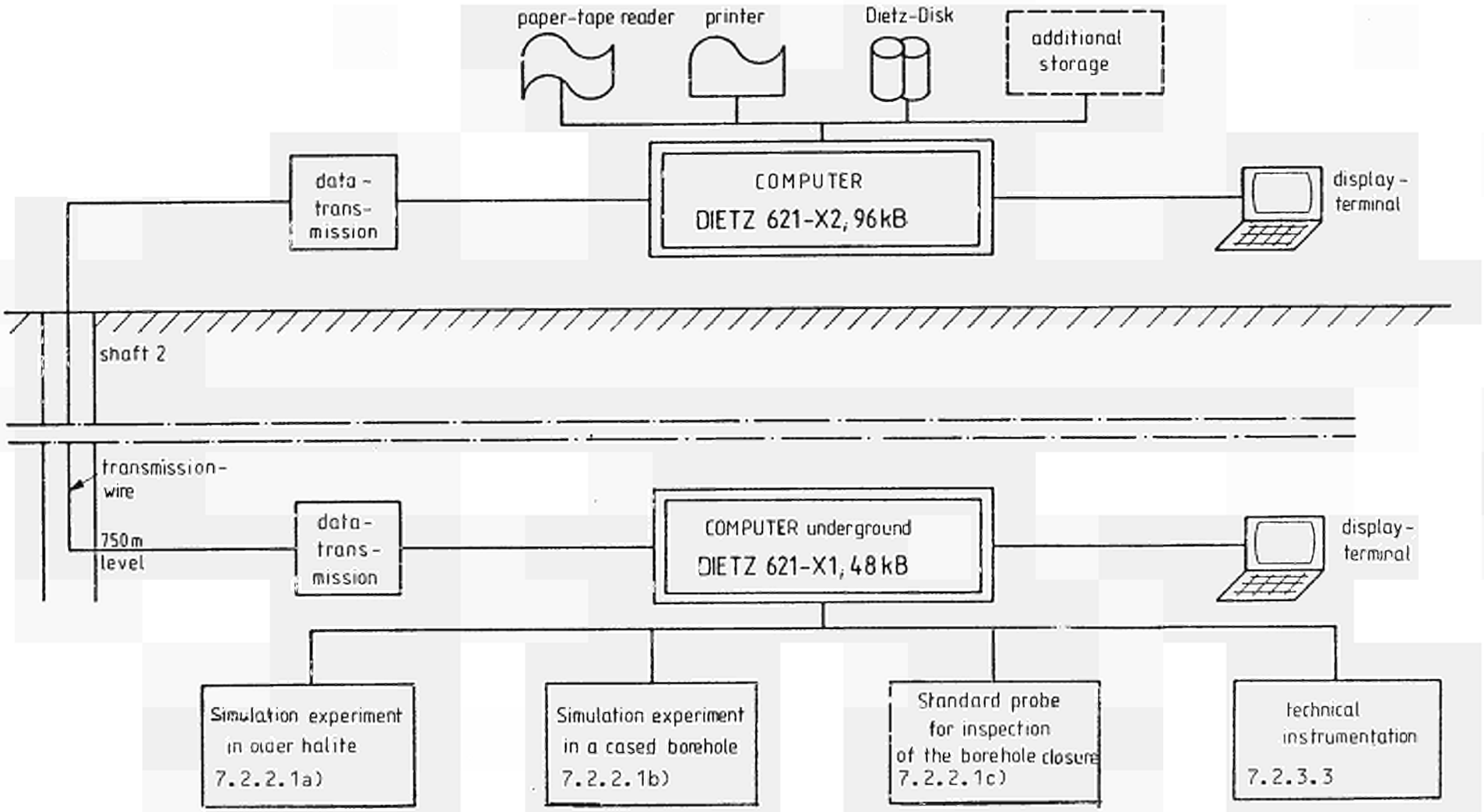


FIGURE 7.2.7 : BLOCK DIAGRAMM OF THE COMPUTER SYSTEM TO BE INSTALLED AT THE ASSE SALT MINE

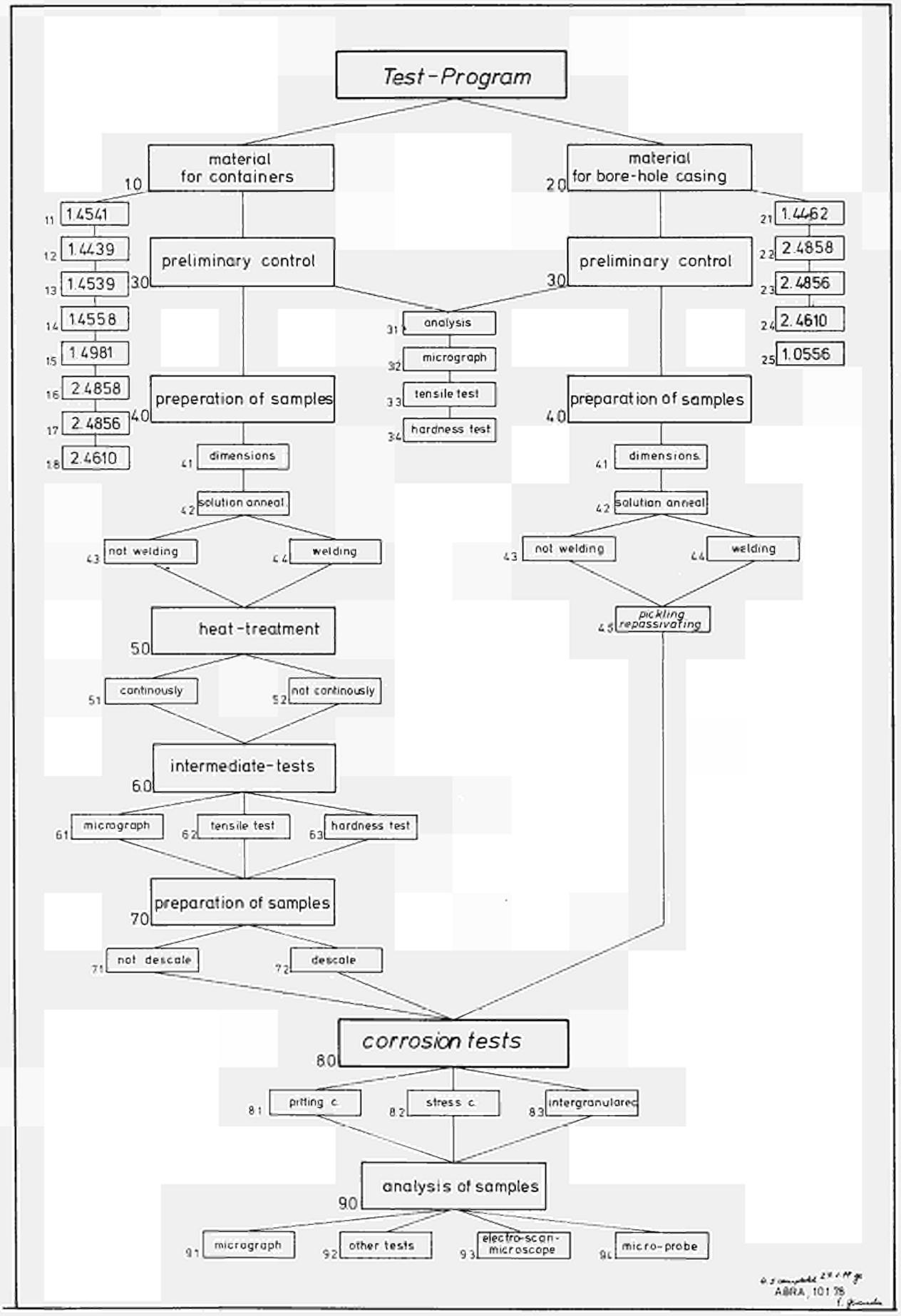


Figure 7.2.8 : Corrosion test programme of borehole casing and waste canister materials

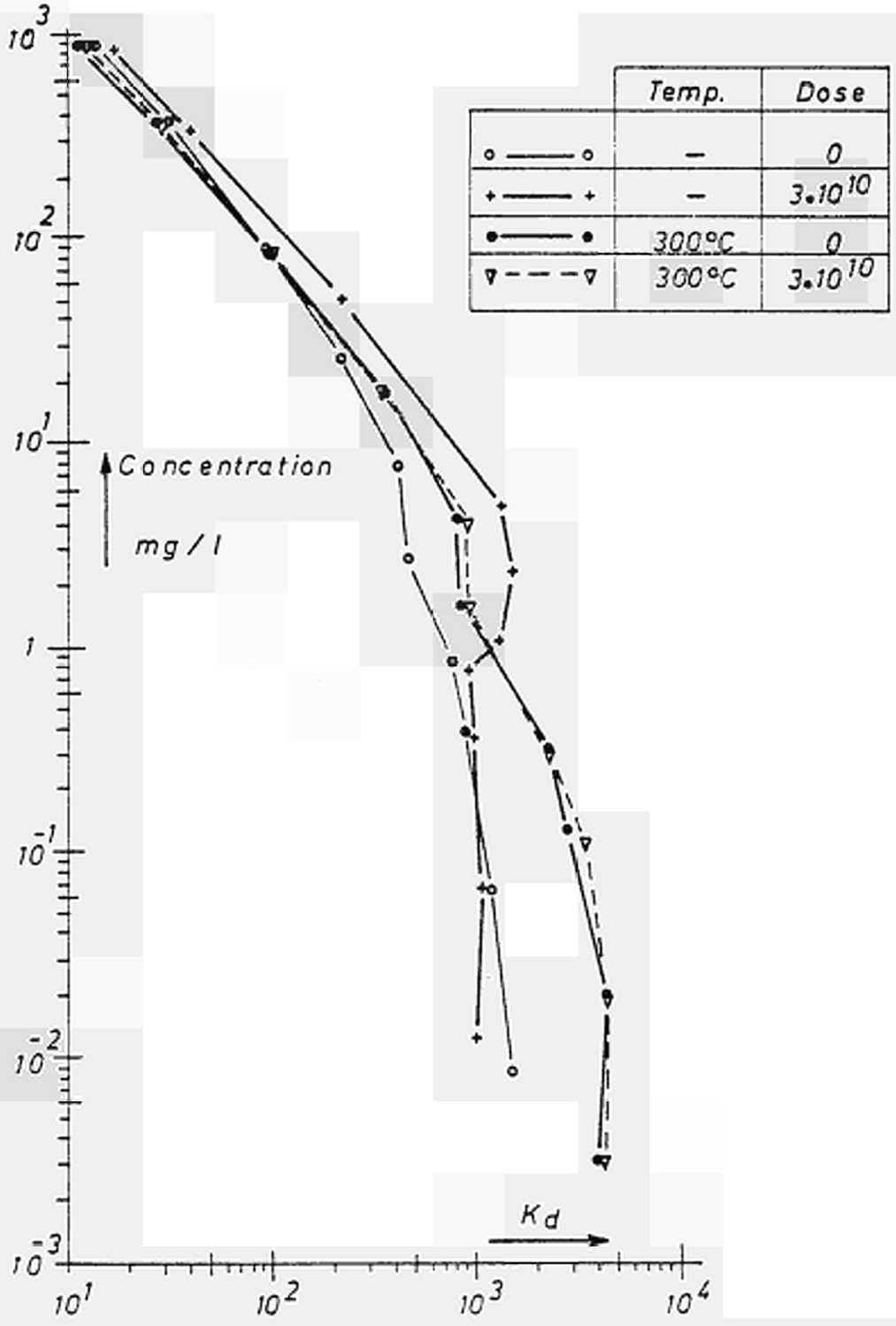
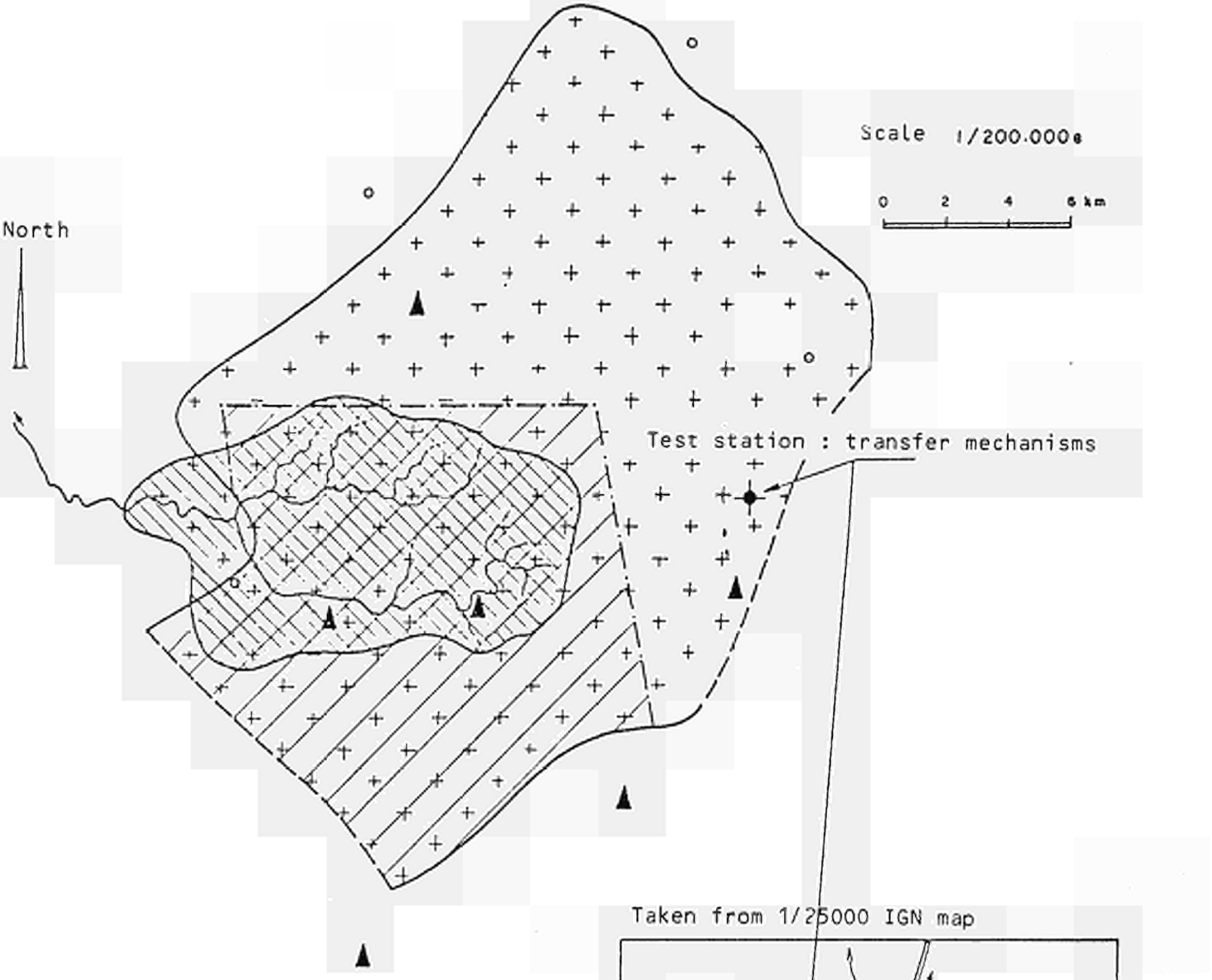


Fig. 7.4.1 : Variation of the distribution coefficient for Eu variable concentration

Influence of temperature and irradiation

(~220 m clay samples)



Area of catalogued existing wells

Experimental basin : measurements and down-the-hole hammer drilling campaign (see detailed inset)

Granite area studied

Climatological station

Quarry subjected to structural analysis

E 7.6.1 : MASSIF B - HYDROGEOLOGICAL STUDY
LOCATION OF INVESTIGATIONS

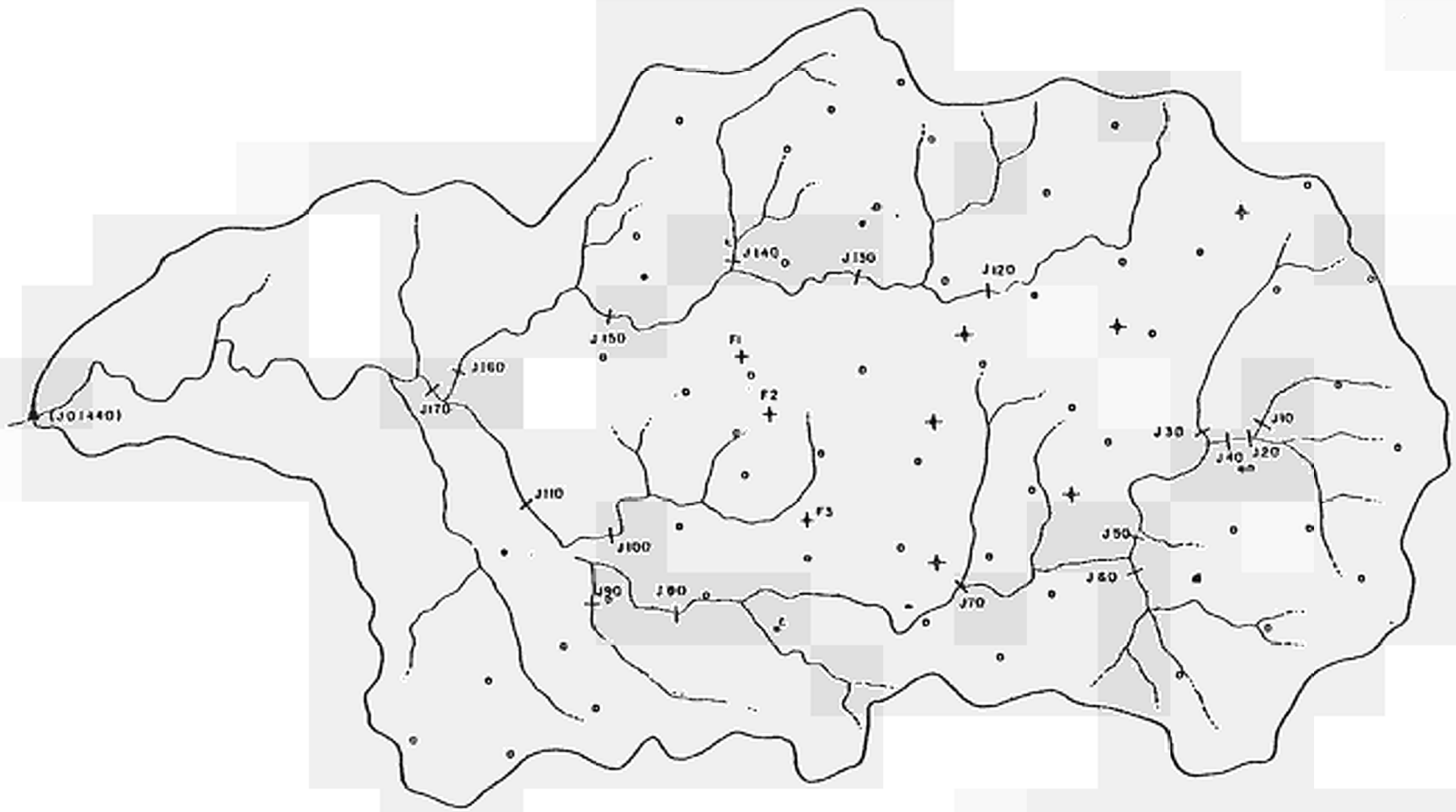
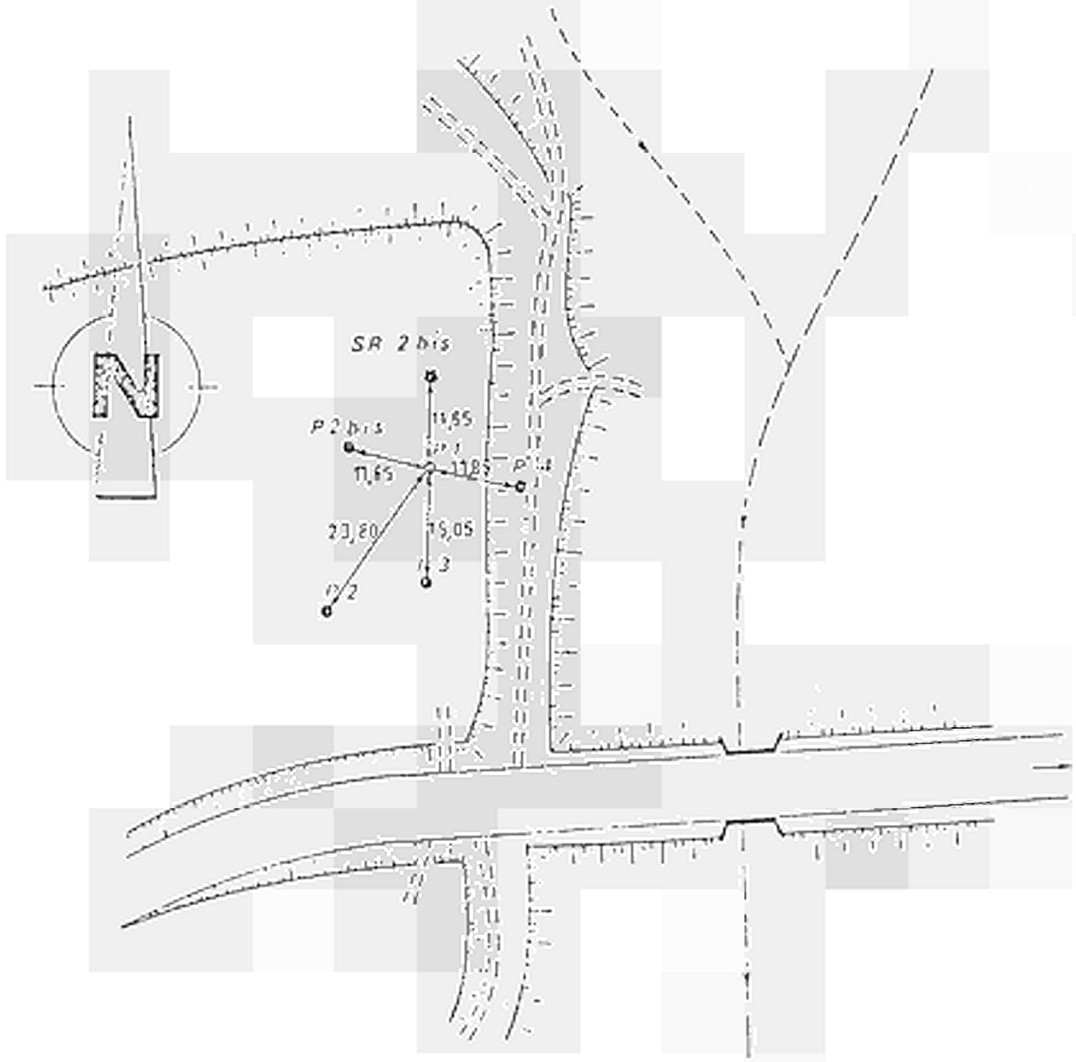


Figure 7.6.2 : Massif B - hydrogeological study - experimental basin

Scale : 1/1 000 environ



- Pneumatic drilling
- Core drilling

Figure 7.6.3 : Sketch of location of exploratory drillings

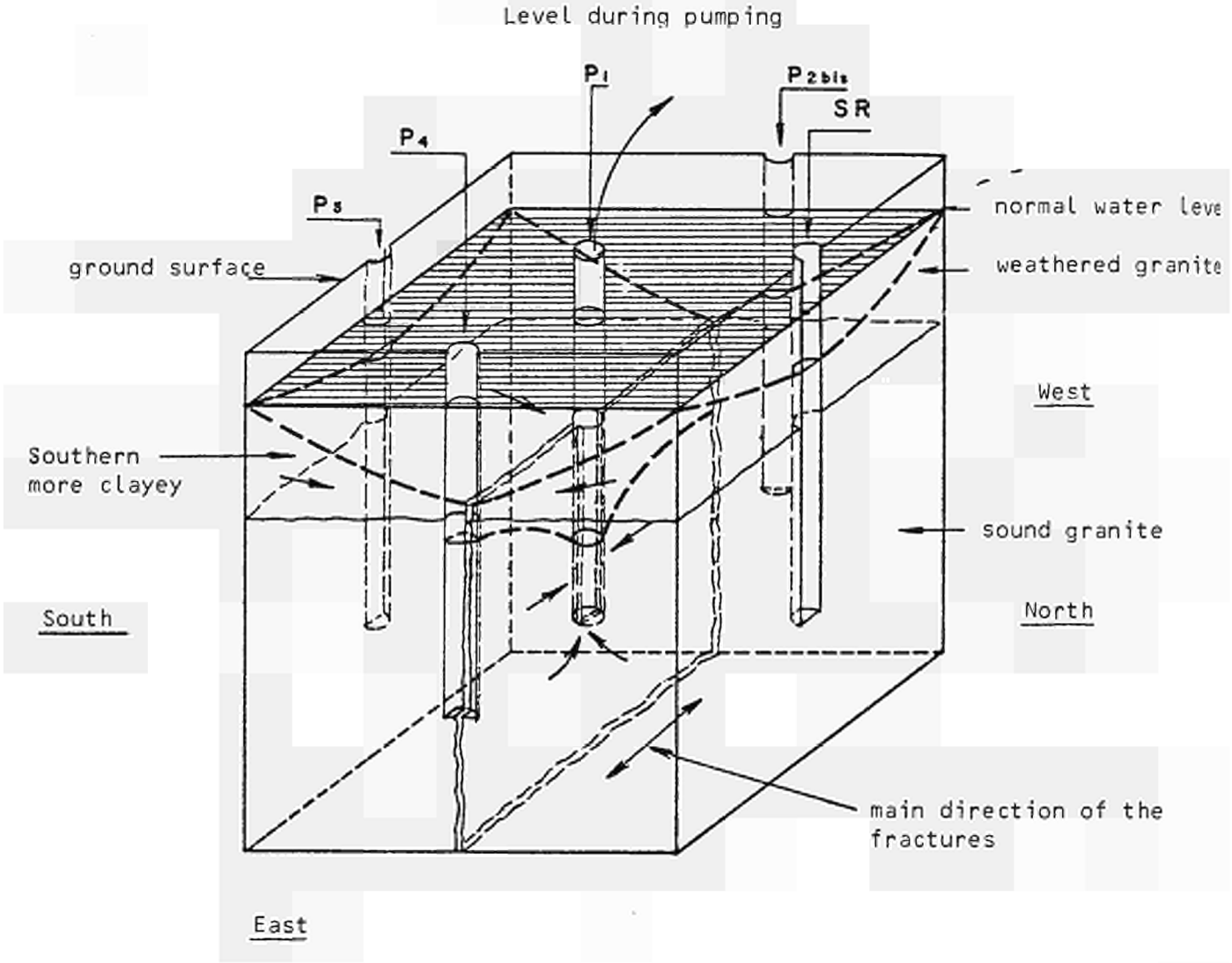


Fig. 7.6.4 : Hydrodynamic diagram of experimental Massif B

Figure 7.6.5 : Chemical composition of the materials

	Illite %	Sépiolite %	Silice %	Attapulgite %	Clinoptilolite %	Magadiite %	Bentonite %	Kaolinite %	Vermiculite %
Si O2	51,40	45,40	99,70	55,20	58,60	73,60	49,60	45,80	40
Al2O3	19,30	0,70	0,22	11,30	15,20	0,39	16,35	37,20	10,5
FE2O3	6,60	0,45	0,14	4,40	3,75	0,15	4,70	1,50	5
FEO	< 0,10	< 0,10	< 0,10	< 0,10	< 0,10	ND	ND	ND	1
TiO2	0,56	0,11	0,06	0,59	0,15	0,02	0,32	0,29	1,5
MnO	0,05	0,01	0,01	< 0,01	0,01	< 0,01	0,05	< 0,01	-
CaO	1,40	1,25	0,15	1,00	3,30	0,42	4,00	0,24	1
MgO	3,60	19,20	ND	4,90	1,25	0,22	4,65	0,40	27
Na2O	0,40	0,06	0,02	0,06	0,85	7,40	2,65	0,05	-
K2O	8,00	0,15	0,03	1,70	3,05	0,35	1,20	0,07	5
P2O5	0,38	0,04	0,14	0,04	0,09	0,02	0,05	0,05	-
H2O⁻	2,70	22,75	0,05	13,00	3,95	9,75	6,15	0,30	9
H2O⁺	5,65	8,90	ND	8,25	9,60	5,30	5,45	14,10	-
Ci CO2						0,13			

Radioactive elements / Absorbent materials	Caesium Cs	Strontium Sr	Zirconium Zr	Tecnetium Tc	Tin Sn	Cerium Ce	Samarium Sm	Neptunium Np	Plutonium Pu	Americium Am	Iodine I
Quartz	+ / **	+ / *	+ / ***	+ / *	+ / ***	+ / ***	+ / ***	+ / *	+ / ***	+ / ***	
Magadiite	+ / **	-	-	-	-	+ / ***	-	-	-	-	
Bentonite	+ / ***	+ / ***	+ / ****	+ / *	+ / ****	+ / ****	+ / ****	+ / ****	+ / ****	+ / ****	
Clinoptilolite	+ / ****	+ / ***	+ / ****	+ / **	+ / ****	+ / ****	+ / ****	+ / **	+ / ***	+ / ****	
Kaolinite	+ / ***	+ / ***	+ / ***	+ / **	+ / ****	+ / ****	+ / ****	+ / **	+ / ***	+ / ***	
Sépiolite	+ / ****	+ / ***	+ / ****	+ / *	+ / ****	+ / ****	+ / ****	+ / **	+ / ****	+ / ****	
Attapulgite	+ / ***	+ / ***	+ / ****	+ / *	+ / ****	+ / ***	+ / ****	+ / **	+ / ****	+ / ****	
Illite	+ / ***	+ / ***	+ / ****	+ / *	+ / *****	+ / ****	+ / ****	+ / **	+ / ****	+ / ****	
Bauxite	+ / ***	+ / **	+ / *****	+ / **	+ / ****	+ / *****	+ / *****	+ / ***	+ / ****	+ / ****	
Vermiculite	+ / ****	+ / ***	+ / ***	+ / *	+ / ****	+ / ****		+ / **	+ / ****	+ / ****	
Zirconium	+ / *	+ / *	+ / *****	+ / *	-	-	+ / ****	-	-	-	-
Zéolite 13 X	+ / ***	+ / *	-	-	-	-	+ / ****	+ / **	+ / ***	+ / ****	-

\+ completed
\- abandoned

*	$0 < K_d < 10$
**	$10 < K_d < 10^2$
***	$10^2 < K_d < 10^4$
****	$10^4 < K_d < 10^6$
*****	$K_d > 10^6$

Fiture 7.6.6. : Recapitulation of Kd values obtained at equilibrium pH values of material-solution in experimental conditions (buffer pH)

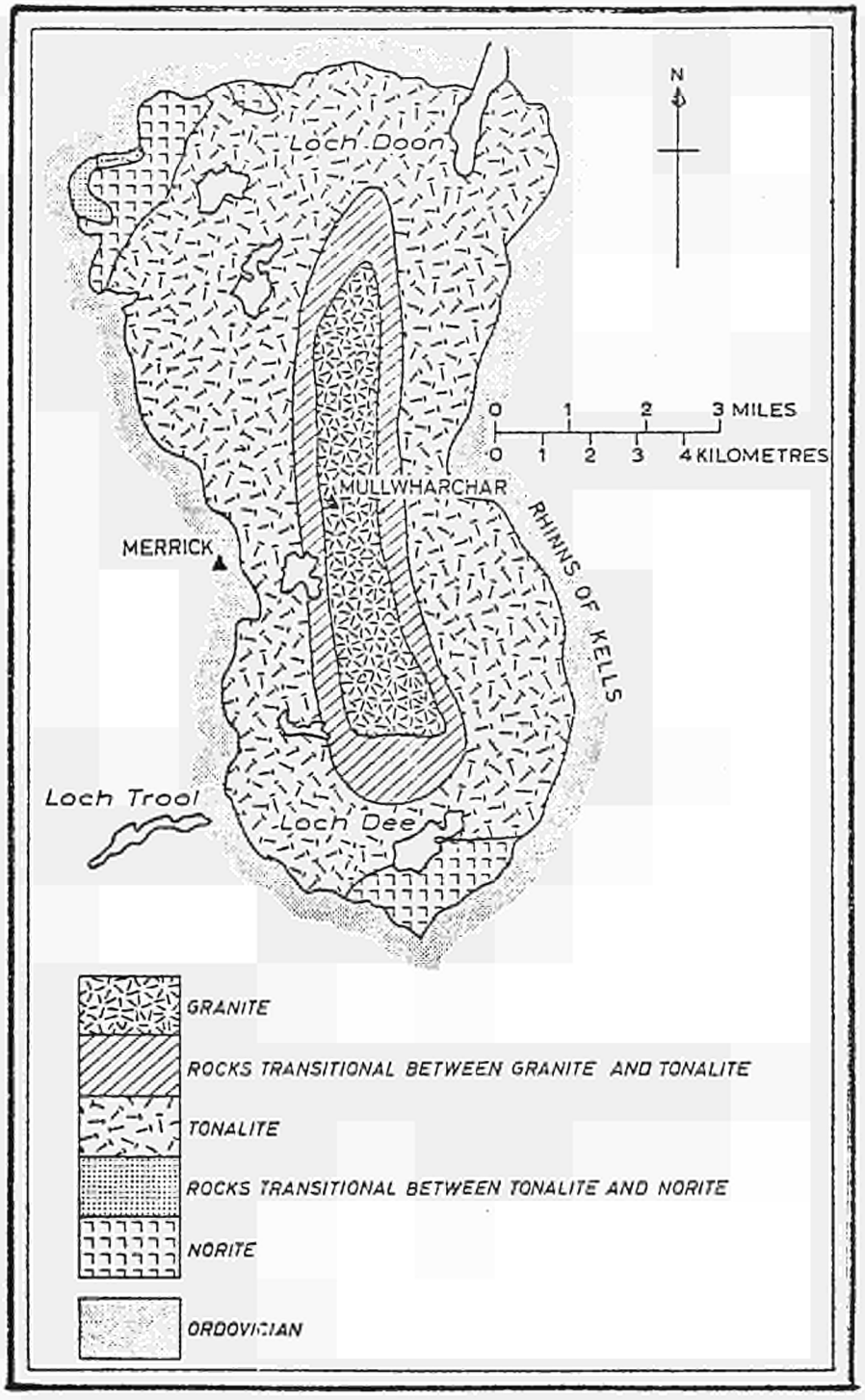


Figure 7.7.1 : Outline geological map of the Loch Doon Intrusion

PROPOSED EXPERIMENTAL PROGRAMME

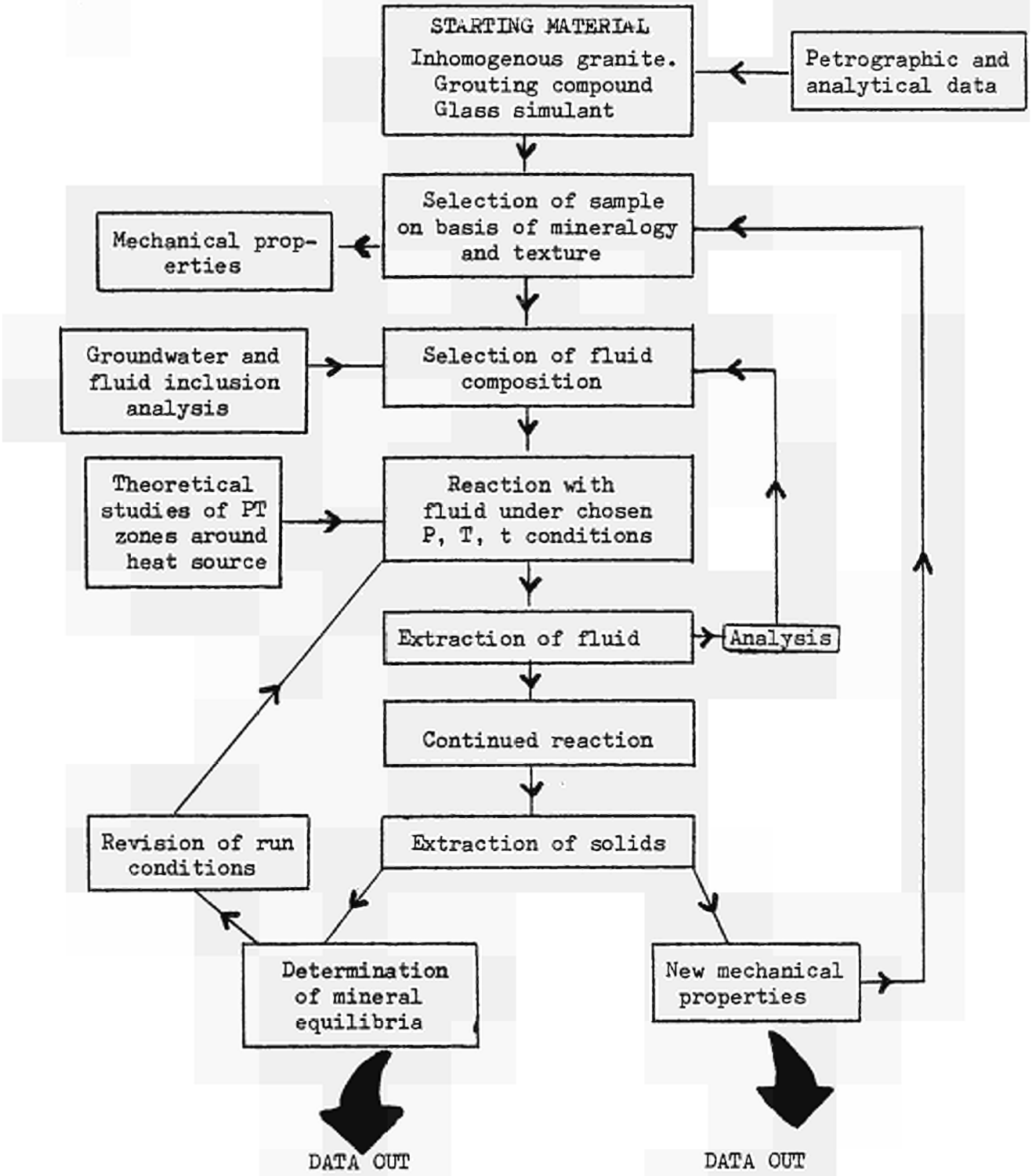


Figure 7.7.2 : Experimental programme to investigate the interaction between hydrothermal fluids and rock materials at elevated temperature and pressure

Characteristic	Suggested Logging Technique
Fractures - in hole (surface)	Single point resistivity Caliper Logs Sonic Amplitude Borehole Television Acoustic Televiewer
Fractures - rock matrix (volume)	Sonic Amplitude Shrimp - possible development
Primary Porosity and Interstitial water quality	Calibrated Resistivity Neutron Probe Gamma-Gamma logs
Lithology	Electric Logs - resistivity Radiation Logs - gamma and neutron Caliper Logs
Moisture Content above water level	Neutron Logs - calibrated
Well Water Flow	Tracing techniques with - Borehole Television - Radiotracers - Conductivity - Thermal Probes - Flow Meter
Clay Fill of Fractures	Gamma Log

Fig. 7.7.3 - Equipment to determine hydraulic properties of poorly permeable fractured media

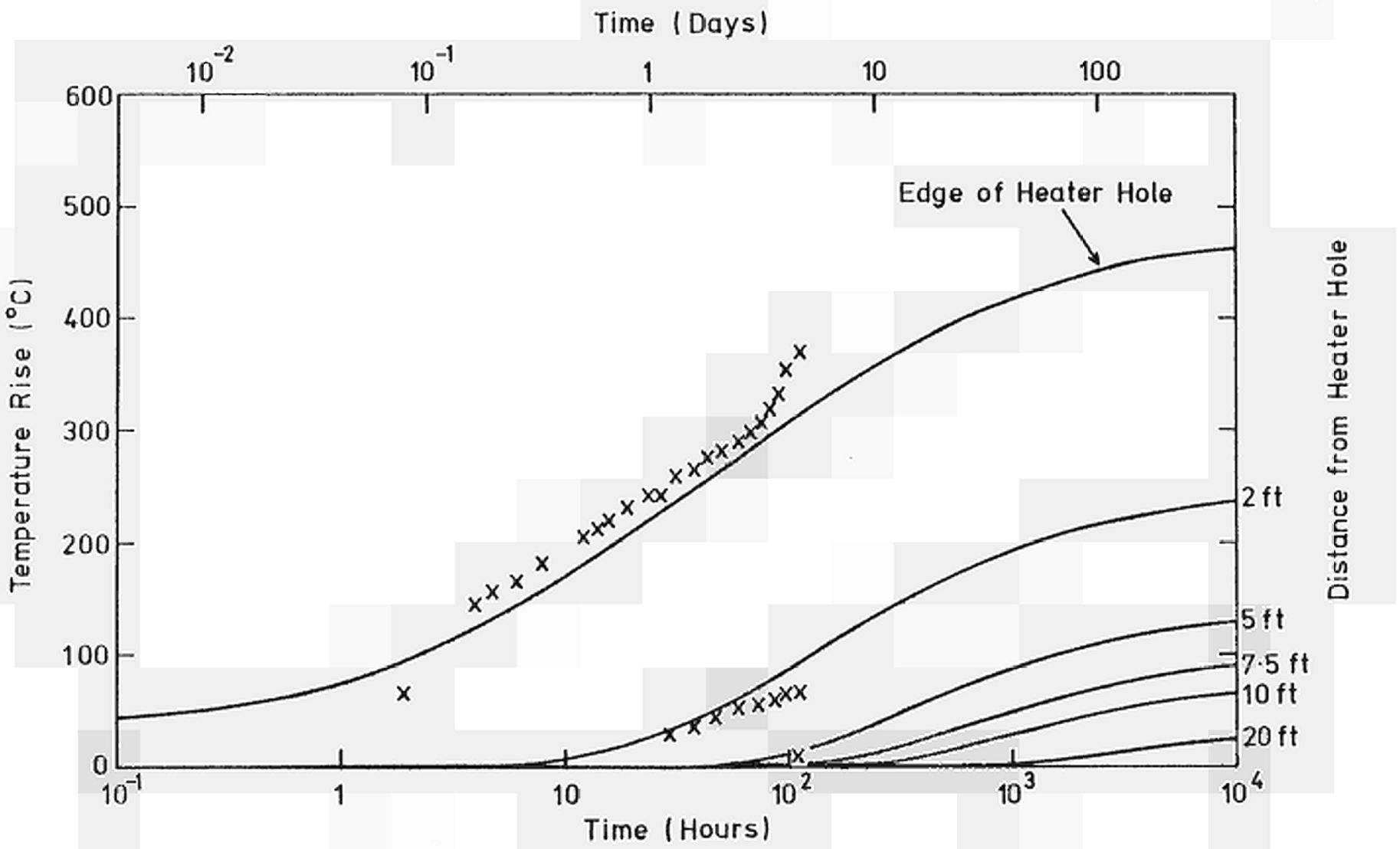


Figure 7.7.4 : Heating experiment in granite (Cornwall)
(Temperature Rise vs Time)

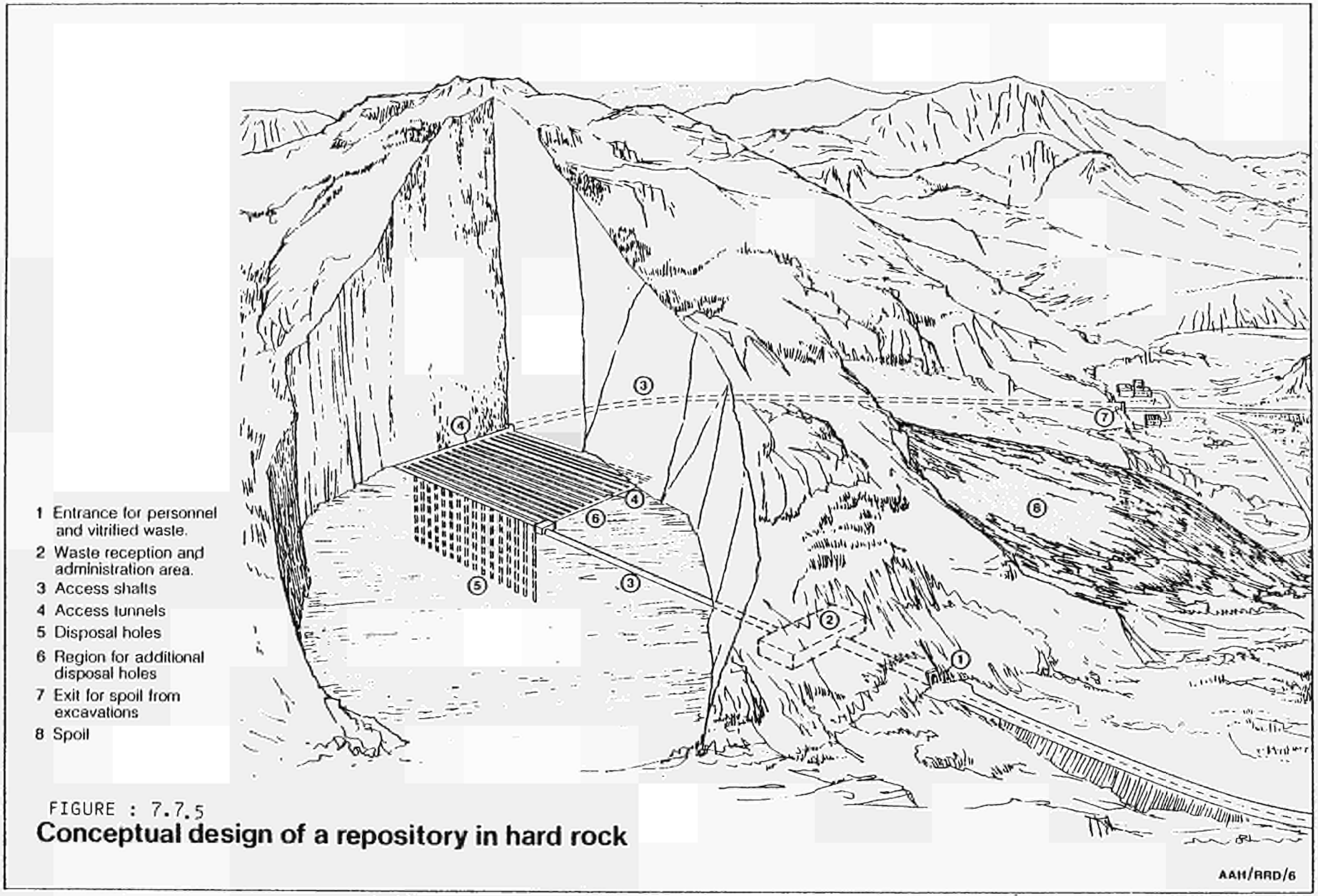


FIGURE : 7.7.5

Conceptual design of a repository in hard rock

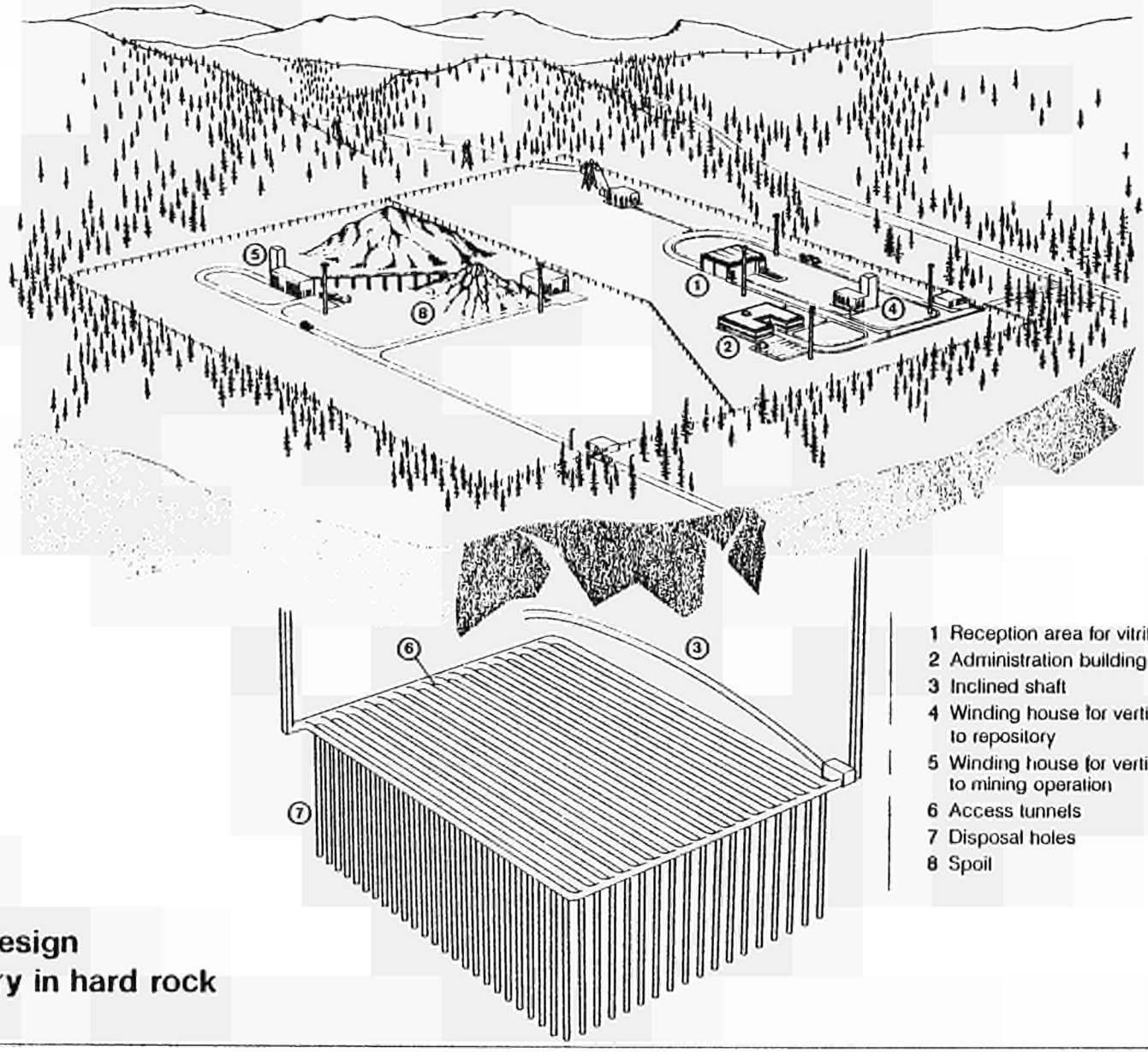


FIGURE : 7.7.6

Conceptual design of a repository in hard rock

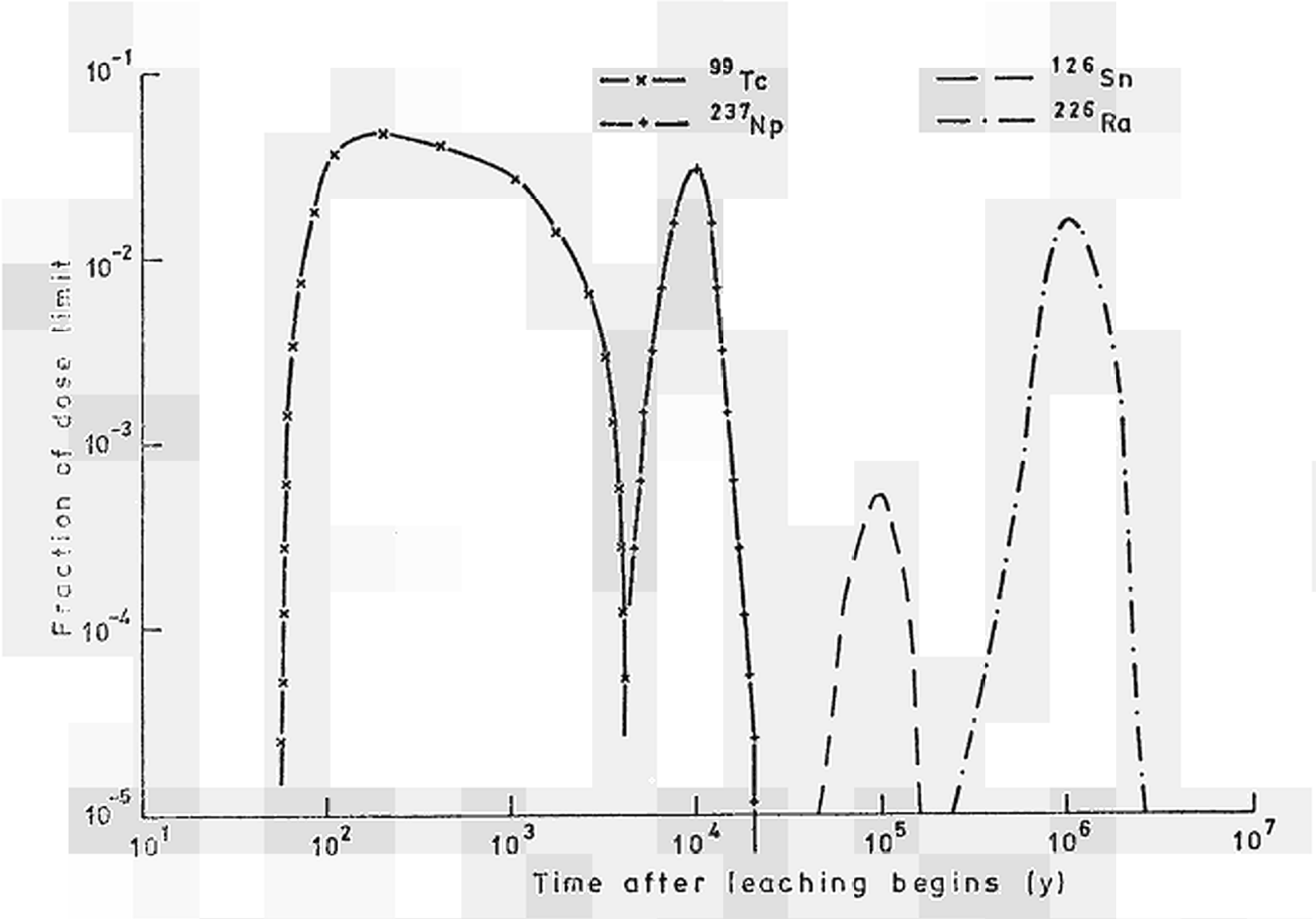


FIGURE 7.7.7 : Doses to individuals from drinking water

8 STORAGE OF GASEOUS WASTE

8.1. General Introduction

With the increase in nuclear power it will become necessary to retain certain radioactive nuclides released during fuel reprocessing (table 8.1).

Table 8.1 : Gaseous and volatile radionuclides in spent LWR fuel elements irradiated to 33 GWD/MT and cooled for 1 year.

Element : Radioisotope	Half-life in years	Amounts per metric ton of fuel		Arisings from 1400 metric tons of reprocessed fuel	
		Volume or Weight	Activity (Heat decay)	Volume or Weight	Activity
Krypton (Kr-85:7 %)	10.8	100 l stp	10.6 kCi (0.18 w/l stp)	140 m^3 stp	15 MCi
Iodine (I-129:85%)	$1.6 \cdot 10^7$	280 g	40 mCi	390 kg	55 Ci
Hydrogen (H-3:~100%)	12.4	~0.6 l stp	700 Ci	0.8 m^3 stp	~1 MCi

A programme to improve or develop alternative methods of conditioning and storing these radionuclides was introduced in the last annual report (EUR 5749 e,f - Chapter 7.1)

8.2. Krypton Incorporation in a Metallic Matrix (UKAEA, Harwell)

The objective of this study is to design, construct and operate a half scale pilot plant to provide data for an industrial plant and to assess the process and product.

The process essentially consists of the implantation of the gas as ion into a metal by means of a glow discharge (Fig. 8.2.1 A) and then sealing it by coating it with metal sputtered from the other electrode (Fig. 8.2.1 B). By repeating the process a thick layer of deposit is built up containing about 170 litres of Krypton per liter of metal.

A schematic representation of a plant unit during operation and in long term storage is given in figure 8.2.2.

8.2.1. Research Programme

The reference gas to be stored is the Krypton which is removed - typically by cryogenic distillation - from the dissolver off-gas of a reprocessing plant.

Inactive krypton will be used except where specified otherwise.

8.2.1.1. Environmental, Safety and Conceptual Process Design Studies

A study will be made of the environmental characteristics and safety of the matrix storage method compared with alternative methods of storing radioactive krypton from industrial fuel reprocessing plants. In addition a conceptual process design study of the full scale industrial plant for incorporating the krypton in the matrix will be provided. This will include estimates of the optimum design parameters; the power consumption and other cost information; plant control and operational characteristics; waste generation; and the extent to which the impurities have to be removed from the krypton.

These studies will provide a basis for the choice of the matrix material and of the design parameters for the pilot plant. The studies will be revised, where necessary, to take into account the results from operation of the pilot plant.

8.2.1.2. Preliminary Studies

These studies should provide detailed information required for the design of the pilot plant. The work will include :

- The design, construction and testing of a prototype rapid switch system for reversing the current since this rapid switching involves some novel problems;
- Tests on candidate metals to select the optimum one for the metal matrix. Inert krypton filled layers of selected materials will be deposited. The gas concentration and stability, and pumping efficiency (litres/kWh) will be measured. The deposits will be examined using appropriate metallurgical techniques;
- Investigation of detailed design features for the process vessel;
- Investigation of process parameters (e.g. voltage, switch frequency and current density) beyond the range so far covered;

- Tests to determine the effect of small amounts of other gases in the krypton, for example, xenon, air, hydrogen, water, etc.

The equipment which will be used for these studies is existing and available at Harwell.

8.2.1.3. Design, Construction and Commissioning of a Pilot Plant

A pilot plant of nominally 1/2 full size (vessel size typically 20 cm diameter, 30 cm long) will be designed, constructed and commissioned. The major components of this plant will be :

- Power supply (i.e. transformer and associated control system) capable of providing 50 kVA at variable voltages up to 10 kV;
- Rapid switch system for reversing the current;
- Process vessels. It will be possible to connect various process vessels for different experiments;
- Associated vacuum, cooling water, control and measurement systems.

8.2.1.4. Operation of the Pilot Plant

Design and operational information will be obtained using the pilot plant with the following objectives :

- The general principles of the process will be demonstrated;
- The optimum operating conditions of the plant will be determined;
- The efficiency of the plant will be determined (in terms of litres of gas stored per kWh and useful volume utilisation);
- The operating characteristics and control features of the plant will be investigated;
- Reliable operator free operation under automatic control for long periods will be demonstrated;
- Possible causes of breakdown will be identified;
- The design limitations of the plant will be determined (e.g. limiting thickness of matrix, electrode spacing, current density);
- A special test will be carried out with fully active krypton to identify any effect on operational performance.

8.2.1.5. Testing of Krypton Filled Matrix Material

Thick specimens of Krypton filled matrix will be produced using the pilot plant. This material will be characterised and tested to establish its performance over the required long periods of storage. Trace active krypton will be used where required for the measurements. The following works will in particular be carried out :

- The matrix will be examined using appropriate metallurgical techniques such as optical, scanning and transmission microscopy;
- The thermal stability and krypton release versus temperature characteristics of matrix will be measured by an established technique of heating the specimens in a vacuum and measuring the quantity of krypton released;
- The long term stability of the matrix will be investigated by incapsulating samples in evacuated containers and determining the quantity of any krypton released after storage at different temperatures;
- The radiation stability of the matrix will be investigated by bombardment with electrons of the appropriate energy;
- The leach rate of the matrix will be measured using suitable techniques including the Soxhlet apparatus;
- The effect of rubidium generated by the decay of krypton 85 in the matrix will be investigated by injecting samples of the matrix with rubidium ions from an ion accelerator and observing the results at various temperatures using suitable techniques. For this the Harwell heavy ion accelerator linked to a 200 kV electron microscope in which the effects of bombardment can be simultaneously viewed, or other high energy implantation facilities will be used;

The equipment which is required for these studies is existing and available at Harwell.
A schematic representation of a plant unit during operation and in long term storage is given in figure 8.2.2.

Table 8.2 : Timetable of research programme (from July 1977 to December 1979)

Point of programme	1978							1979			
	III	IV	I	II	III	IV	I	II	III	IV	
Environmental, safety and process design study	———			———						———	
Preliminary study	———			———							
Construction of a pilot plant	———			———							
Operation of a pilot plant				———							
Testing of the conditioned material					———						

8.3. Krypton Storage in Pressurized Cylinders (KFA, Jülich)

The object of this research is to develop the concept of a long term storage of krypton in pressurized cylinders in an engineered structure and to evaluate its safety and economic aspects.

8.3.1. Research Programme

The reference gas to be stored is the krypton removed by cryogenic distillation from the dissolver off gas of a reprocessing plant with a capacity of 1400 t/a.

8.3.1.1. Literature Review and Basic Data

Existing literature on storage of radioactive krypton in pressurized containers will be reviewed in order to constitute the starting-point of the action. Main design data of the storage system will be determined. The following subassemblies will be considered.:

- Storage cylinder;
- Filling station;
- Transport equipment;
- Storage building including the required cooling system, ventilation, shielding, etc.

Arisings and properties of the krypton as well as the principal characteristics and conceptual criteria for the different parts of the system will also be evaluated, taking into account the licensing guidelines.

8.3.1.2. Laboratory Study

In case of insufficient literature data the following data will be experimentally determined :

a) Heat transfer in the system Krypton-storage cylinder-coolant

Storage cylinder models with simulated heat sources will be used for experimentation.

b) Corrosion of storage cylinder

The interaction of rubidium, the krypton decay product, with the container material, taking into account the influence of radiation and gas impurities, will be studied and results extrapolated to the required storage term.

c) Krypton immobilization in an absorber

If suitable, the krypton immobilization in an absorber (Charcoal, M.S.) will be studied by using small amounts of radioactive krypton.

8.3.1.3. Design Study of the Storage Facility

On the basis of obtained results a concept of the storage facility will be elaborated.

8.3.1.4. Safety and Economic Analysis

A typical accident for each part of the system will be considered and the consequences will be evaluated (including the casual container failure).

Capital and operating costs will be evaluated (DM/kg uranium) and compared to alternative storage concepts.

8.3.2. Schedule of the Research Programme

Table 8.3 : Timetable of the research programme (from July 1977 to December 1979)

Point of the programme	1977		1978				1979			
	III	IV	I	II	III	IV	I	II	III	IV
Literature review	———									
Laboratory study			———	———	———	———	———			
Storage study				———	———	———	———			
Safety analysis							———	———		
Economic study									———	———

8.4. Storage of Charcoal Absorbed Krypton in Pressurized Cylinders (CEN/SCK, Mol)

The incorporation in activated charcoal of the radioactive krypton to be stored in pressurized cylinders, may bring some advantages such as internal pressure reduction and attenuation of gas release in case of container failure.

8.4.1. Research Programme

8.4.1.1. Characterization of the Activated Charcoals

The following measurements will be taken on different types of charcoal :

- Porosity distribution (micro and macro-pores) by nitrogen absorption and by mercury porosimetry;
- Density (in mercury).

Charcoals (3-4 types) will be selected to determine the krypton absorption rates at temperatures and pressures from -10°C to +50°C and from 0 to 900 mm of Hg respectively.

8.4.1.2. Densification of Activated Charcoal Beds

Optimizing of preparation techniques by vibration of single or mixed phases such as :

- Granulated charcoal/powdered charcoal;
- Granulated charcoal/metal powder;
- Granulated charcoal/powdered graphite.

The chosen charcoal bed will be used for the thermal conductivity measurements.

8.4.1.3. Heat Transfer

Measurements will be carried out on the different dried charcoal beds using nitrogen or krypton atmosphere. An appropriate equipment will be assembled, consisting of a column (diameter : 95 mm) with a central heating element and thermo-couples.

A mathematical model will be established to calculate the temperatures, pressures and krypton loads in cylinders having diameters of 5 and 20 cm. These calculations will be performed on the most favourable combination of thermal conductivity and adsorption capacity of charcoal beds.

8.4.1.4. Influence of Rubidium

Rubidium, which is the decay product of krypton-85, may cause a corrosion problem of the container. An experiment aimed at assessing this problem in presence of charcoal, will be conceived by comparing the actual vapour

pressure of the charcoal absorbed rubidium to that of the free metal. An attempt will be also made to show the rubidium migration from the container wall towards the charcoal.

Table 8.4 : Timetable of the research programme (from January 1977 to June 1978)

Point of the programme	1977				1978	
	I	II	III	IV	I	II
Charcoal characterization	———	———	———			
Densification of charcoal beds			———		———	
Heat transfer	———	———	———	———	———	
Influence of rubidium				———	———	———

8.5. Feasibility of Krypton Sea Disposal (KFA, Jülich and ECN, Petten)

A theoretical study has to be carried out on the whole concept of the sea dumping of krypton in pressurized containers as a method of disposal. KFA-Jülich and ECN-Petten proposals have harmonized their programmes to avoid, as far as possible, duplication of work. KFA will concentrate its efforts on the krypton handling and treatment before dumping, using conventional cylinders with re-usable shielding. ECN will study radiological, legal and logistic aspects of the sea disposal itself and different types of containers with integrated shielding.

8.5.1. KFA-Jülich Research Programme

The object of this research action is an analysis on the problems related to sea disposal of krypton.

8.5.1.1. Characterization of the Gas to be Disposed of

On the basis of the reference data it will be evaluated :

- Krypton and rubidium contents in the container as a function of time;
- Radiation;
- Heat generation;
- Rubidium problems (corrosion, chemical reactions).

8.5.1.2. Container Characteristics

A stainless steel cylinder with a volume of not more than 50 liters is assumed to contain the krypton. The cylinder, which has to be hermetically sealed, may be provided, with a special valve. The relevant parameters are :

- Radioactivity content;
- Temperature and pressure limits;
- Material problems (internal and external corrosion, time of storage).

8.5.1,3. Means of Transportation

Specifications will be identified for the transport of gas cylinders from a planned reprocessing plant in Germany to dumping area.

The transport of a gas-tight-type B container will be examined and particular attention will be paid to the internal temperature reached in case of a fire accident.

The following important aspects will be investigated :

- Single and collective containers;
- Accident conditions;
- Heat transfer in normal and accident conditions;
- Shielding.

8.5.1.4. Feasibility and Concept

The whole concept of krypton sea dumping and its feasibility will be defined. The important aspects are :

- Filling of the pressurized cylinders at the cryogenic distillation plant;
- Sealing and leak testing of filled cylinders;
- Interim storage of containers;
- Transport to the harbour;
- Transport by ship and dumping operation.

8.5.1.5. Components and Management Steps

Main emphasis of the study will be on components design and on safety analysis.

Fabrication and materials problems will be identified and the operation of the components behaviour during operation in normal and accident conditions will be evaluated.

The following aspects will be investigated in the safety analysis :

- The pressurized cylinders will be designed to relevant German standards making special allowance for the high radioactive gas;
- Filling, scaling and leak detection will be provided in the gas treatment plant;
- The heat evacuation and a continuous leak monitoring of the containers will be assured during the interim storage and the optimum storage period will be evaluated;
- According to the "Railway regulations" and the "European convention for transport by land of dangerous goods", a reusable transport container (type B) will be required;
- A ship specially equipped with decay heat removal system and a rig for the dumping will be needed.

8.5.1.6. Behaviour of Krypton after Sea Dumping

In consideration of the ECN-Petten programme on the subject (see Chap. 8.5.2) this study will be limited to the krypton hydrate formation and in particular :

- Hydrate formation as function of pressure and sea-water temperature;
- Influence of the decay heat on stability of the hydrates;
- Diffusion of the hydrates.

8.5.1.7. Cost Evaluation

A cost estimate of the developed concept will be prepared.

8.5.1.8. Accidents

The consequences of accident in the system for krypton sea disposal will be analysed and evaluated as individual dose under conservative assumptions. Accident related to all relevant operations will be analysed.

8.5.1.9. Legal Problems

As this aspect will be mainly treated by ECN-Petten, only specific problems arising in Germany will be examined : for instance considerations in view of the extension of "Regulations on pressurized gas" and of "Technical principles" to handling self-heating radioactive gases.

Table 8.5 : Schedule for the programme execution (from January 1978 to December 1979)

Point of the programme	1978				1979			
	I	II	III	IV	I	II	III	IV
Container and transport	———							
Feasibility and concept	———	———	———	———				
Components and management steps			———	———	———	———	———	
Krypton behaviour after dumping								———
Safety and legal aspects								———

8.5.2. ECN-Petten Research Programme

8.5.2.1. Design Study of the Container

The krypton container will be designed according to the rules and requirements of the IAEA Regulations for the Safe Transport of Radioactive Materials, 1973 Revised Edition. One of the consequences is the application of the double-containment concept. The container will be designed in such a way that in the unlikely event of the inner vessel failure the outer vessel can withstand the pressure of the krypton during and after the test period. The shielding will form an integral part of the outer vessel and will consequently be dumped together with the other parts of the container. Such a container can easily be stored, transported and dumped at sea at any suitable moment without special provisions.

8.5.2.2. Feasibility Study

This study will be limited to cylindrical and spherical containers with 10 and 100 litres capacity and varying filling mass of krypton. The following aspects will be considered :

- Temperature distribution : Apart from the temperature distribution in the stationary situation, special attention will be given to the transient situation during the fire test;
- Strength : The following loads will be considered : internal gas pressure, especially during the fire test, hydrostatic pressure on the outer vessel after sea dumping, temperature induced stresses, impact in case of the drop tests;

- Filling procedure and leak-tight closure of the container
- Leak detection

8.5.2.3. Design of Selected Concepts

Not more than four concepts will be selected for the design study. The following aspects will receive particular attention :

- Choice of material;
- Strength under normal and accident (drop and fire tests) conditions, possible contribution of shielding to strength;
- Heat dissipation outwards;
- Heat inflow during fire;
- Filling and sealing procedure;
- Fabrication technique and assembly procedure including the required shielding;

The amounts of radioactive krypton to be disposed of till the year 2000 will be estimated, on the basis of the forecast reprocessing capacity in the European Community.

8.5.2.4. Sea Disposal of Radioactive Krypton

All stages of the sea disposal operation will be studied, both when using a container with integrated shielding, or when applying a conventional pressurized cylinder with re-usable shielding.

8.5.2.5. Interim Storage and Overland Transport

Since sea disposal operations are of discontinuous character, interim storage is required. This aspect will be briefly treated considering the different types of containers.

Rail and road transport will be compared for the transport to the harbour from the reprocessing plant and the radiological consequences of a container failure during transport will be briefly considered.

8.5.2.6. Ship Transport and Dumping

Special requirements and their influence on the choice of a ship for sea disposal will be investigated. The problem of heat dissipation and the possible thermal shock at dumping will be examinated. Special attention will be given to a convenient technique for dumping cylinders from their transport containers under radiologically safe conditions.

8.5.2.7. Radiological and Legal Aspects

The dispersion of radioactive krypton released from containers after they have reached the ocean floor will be discussed, taking as a basis the radiological safety assessments for sea disposal of high-level waste as recently published by the British National Radiological Protection Board. The uptake capacity of the Nord Atlantic for radioactive krypton will be established and radiological consequences, in terms of dose commitments to man, will be derived to an indicative scale (a full radiological assessment is not in the scope of this study).

The aim and directives of the London Convention will be treated in short. It will then be examined, on the basis of the assessment described in the previous section, whether sea disposal of large amounts of krypton-85, now prohibited, could be tolerated on the basis of radiological consequences and perhaps advocated in comparison with other disposal techniques.

Table 8.6 : Schedule of the research programme (from May 1978 to December 1979)

Point of the programme	1978			1979			
	II	III	IV	I	II	III	IV
Container design study	———	———	———				
Feasibility and selected concepts				———	———	———	
Radiological aspects	———	———	———				
Legal aspects	———	———	———	———	———	———	———

8.6. Krypton Immobilization by Incapsulation in Zeolites (Nukem, Hanau)

Nukem has submitted a proposal for the following research :

a) Molecular sieve selection on the basis of the highest absorption capacity (micro and macro-pores);

b) Absorption conditions : temperature, pressure and loading times optimization and investigation on influence of molecular sieve pre-treatment;

c) Measurement of absorption rate : pre-treatment influence in storage conditions and at sealing temperature (max. value);

d) Sealing : choice of material (resins, glass) and sealing technique;

e) Overall concept study considering safety;

f) Cost estimate : ecently Nukem has proposed to carry out this research project at KfK.

8.7. Screening Study on Tritium Immobilization (UKAEA, Harwell)

The objective of this study is to survey and assess candidate materials for tritium immobilization. The work will take tritium management strategies that may be adopted in the Community and the results should identify the most promising immobilization methods for further development.

8.7.1. Basic Considerations for the Research Programme

As reprocessing plants produce huge volumes of tritiated water - the high activity waste alone is of the order of 5 m^3 per metric ton uranium - new techniques, resulting in smaller volumes, will have to be applied in order to render incorporation of the tritium in a solid material economically acceptable. The following techniques are in particular envisaged :

- Recycling of the aqueous streams : it has been estimated that the volume of waste water could be reduced by a factor of 10 or more;
- Recycling as above, followed by isotopic separation : the optimum concentration should be evaluated (complete separation would lead to about 0,2 cm^3 of T_2O per metric ton uranium;
- Voltoxidization : it has been estimated that by this modified head-end process the tritium could be confined to the order of 10 litres water per metric ton uranium.

As the tritium concentration, depending on the foregoing options, will influence the relative merits of the candidate materials for immobilization, these options should be separately considered in this study and their cost should be taken into account in the economic comparison of the materials.

8.7.2. Identification of Candidate Materials

8.7.2.1. Theoretical Review

A literature study will cover published papers both on specifying methods proposed for tritium storage (including concrete, calcium, sulfate , organic polymers, molecular sieves, metal hydrides, etc.) and on methods for hydrogen storage.

Parallel thermodynamic studies and solid state criteria will be worked out to identify the most promising classes

of material for hydrogen immobilization, such as hydrides and organic materials; this review may allow the identification of highly stable natural minerals (e.g. hydroxy apatite) for further consideration.

8.7.2.2. Criteria for a Tritium Immobilization Technique

The criteria to be met by a tritium immobilization technique will be reviewed and an estimate made of the volume and tritium concentration of arisings for tritium waste management schemes with and without recycle and isotopic enrichment.

8.7.2.3. Preliminary Appraisal

The information derived from the previous sections will be combined and assessed as follows :

- A preliminary list of candidate materials will be assembled, together with significant criteria which will influence selection;
- Limited flowsheet studies as block diagrams will be made to identify unknown areas needing further study;
- If feasible, an initial screening will be made to reduce the number of candidate materials, so that the remainder can be examined in greater depth.

8.7.3. Investigation of Possible Immobilization Schemes

8.7.3.1. Properties of the Product Solid

The solid containing the tritium must be safe and stable under the possible environmental conditions of transport/storage/disposal. The assessment should cover :

- The general stability of the material - such factors as chemical stability, flammability, thermal stability, vapour pressure and friability;
- The properties of the materials within the proposed storage/disposal environment - for example resistance to leaching;
- A review of possible canning methods;
- Any possible interaction between the immobilizing solid and the container material, within possible storage/disposal environments.

8.7.3.2. Effects of Tritium on the Solid

The presence of tritium may pose special stability problems, even for materials which are apparently satisfactory for hydrogen storage. Following studies will be carried out :

- A review of literature data on isotopic exchange

rates for tritium. This should cover possible exchange rates between tritium in the material used for immobilization and water or water vapour;

- An assessment of the effects of radiolysis
  This should cover both the possible effects of radiation on the stability of the host material and the possible formation and release of "hydrogen" gas;
- Any possible pressure effects caused by hydrogen release as a result of radiolysis and any consequent effects on the stability of the host material;
- Any potential hazard which might arise in the event of storage failure (e.g. the release of dust containing tritium).

8.7.3.3. Comparison of immobilization Processes

The following aspects will be considered in particular in the selection of the processes and materials for tritium incorporation :

- Complexity of the process - is a reasonable production process feasible;
- Recycle requirements - is tritium totally retained during the immobilization process, or is recycle of the off-gases needed (e.g. during setting of cement);
- Secondary wastes - are there any arisings that might cause disposal problems (e.g. toxic materials);
- Problem areas - a limited appraisal of the safety of the process is needed (e.g. are there any explosive off-gases).

8.7.4. Overall appraisal and Recommendations

The information obtained will be assessed, using flowsheet studies where appropriate, in order to analyse the most promising systems and to allow a preliminary economic analysis. The outcome will be :

- A list of promising candidate materials for tritium immobilization;
- A list of the classes of material rejected, with reasons;
- An identification of key areas where the data are uncertain and experimental studies are needed;
- A note of any options for "synenergy" in disposal (e.g. the conversion of Zircaloy hulls for zirconium hydride storage);

Recommendations will be provided on experimental work that is needed to complete the assessment study. They will cover :

- Immobilization materials for which experimental stu-

dies on production and tritium storage should be made. If feasible, recommendations will cover materials to suit the various possible tritium waste management strategies;

- Experiments needed to resolve uncertainties in critical data.

Table 8.7.: Timetable of the research programme (from October 1977 to September 1978)

Point of the programme	1977	1978			
	IV	I	II	III	IV
Candidate materials	———				
Investigation of immobilisation schemes		———	———		
Conclusions and recommendations				———	

8.8. Iodine Incorporation in Low Melting Glass (Le Verre Fluoré - S.a.r.l., Rennes)

Stable fluoride glasses have been obtained in the ZrF_4-ThF_4-BaF_2 systems. They have a sufficient viscosity at the melting temperature of about 600°C to retain iodides and therefore they could be used to condition radioactive iodine for final disposal. The objective of this study is the development of new glass compositions for this purpose and the characterization of the obtained products. The iodine to be conditioned is assumed to be separated by various methods at the reprocessing plant and converted to iodide.

8.8.1. Research Programme

8.8.1.1. Solubility Determination of Alkaline and Alkaline-Earth Iodides in the Existing Glasses

The iodide (NaI, KI, BaI_2) quantities which could be dissolved in the glasses of following systems will be determined :

a) ZrF_4-BaF_2-ThF_4

b) ZrF_4-BaF_2-LaF_3

c) ZrF_4-ThF_4-LaF_3

The relations between iodine concentration and various parameters of product quality will be determined.

8.8.1.2. Research of New Low Melting Glasses

The new glass systems in which barium fluoride will be partially replaced by an alkaline fluoride are :

a) ZrF_4-ThF_4-BaF_2-NaF

b) ZrF_4-ThF_4-BaF_2-KF

c) ZrF_4-LaF_3-BaF_2-NaF

d) ZrF_4-AlF_3-BaF_2-NaF

The thermal characteristics and the solubility limit of the various iodides in these new glasses will be determined.

8.8.1.3. Research of Glasses Having an Iodide as Constituent

In order to achieve the highest iodine concentration in the glass, vitreous phases will be examined in ternary or quaternary diagrams of the following systems in which a component is an alkaline or alkaline-earth iodide :

a) ZrF_4-ThF_4-BaI_2

b) ZrF_4-ThF_4-NaI

c) ZrF_4-ALF_3-NaI

d) ZrF_4-ThF_4-BaF_2-BaI_2

e) ZrF_4-ThF_4-BaF_2-NaI

The results obtained with thorium fluoride will be extrapolated to lanthanum fluoride where ever of interest.

8.8.1.4. Adaptation of the Fabrication Process

To avoid that some iodine escapes during conditioning, the existing process will be modified in such way that the oxy-reduction potential of the glass bed would stabilize iodine as I^- anion.

8.8.1.5. Characterization of Iodine Charged Glasses

Following measurements will be carried out :

a) Physical properties

- vitreous transition temperature at melting;
- relative stability of the glasses (vitrification rate);
- resistance of selected glasses against thermal shock (expansion coefficient).

b) Radiation stability

Radiation stability will be evaluated on samples con-

taining stable iodine exposes to beta, gamma and neutron fluxes.

c) Resistance against corrosion

Iodine charged samples will be tested at different temperatures for their resistance in following media :

- normal humid air and steam;
- neutral aqueous solution;
- acid solution;
- basic solution;
- organic solvents (trichlorethylene and oils).

Table 8.7 : Timetable of the research programme (from January 1978 to June 1979)

Point of the programme	1978				1979	
	I	II	III	IV	I	II
Solubility determination	———					
Research on low-melting glass	———	———	———			
Research on iodine-glass			———	———		
Fabrication process study			———	———	———	
Characterisation of iodide-glass	———	———	———	———	———	———

8.9. Iodine Incorporation in Epoxy-Resins (CEN, Mol)

A research on this method of radioactive iodine conditioning to be carried out by CEN is under consideration.

The main points of the programme are :

- Transformation of the retained iodine into an insoluble compound (AgI, PbI_2, $PbIO_3$, CuI);
- Laboratory tests of the pre-treatment techniques;
- Epoxy-resin coating technique;
- Stability and leach tests of the conditioned product in various media (clay, salt, etc.) using I-129;

The detailed research programme has yet to be elaborated.

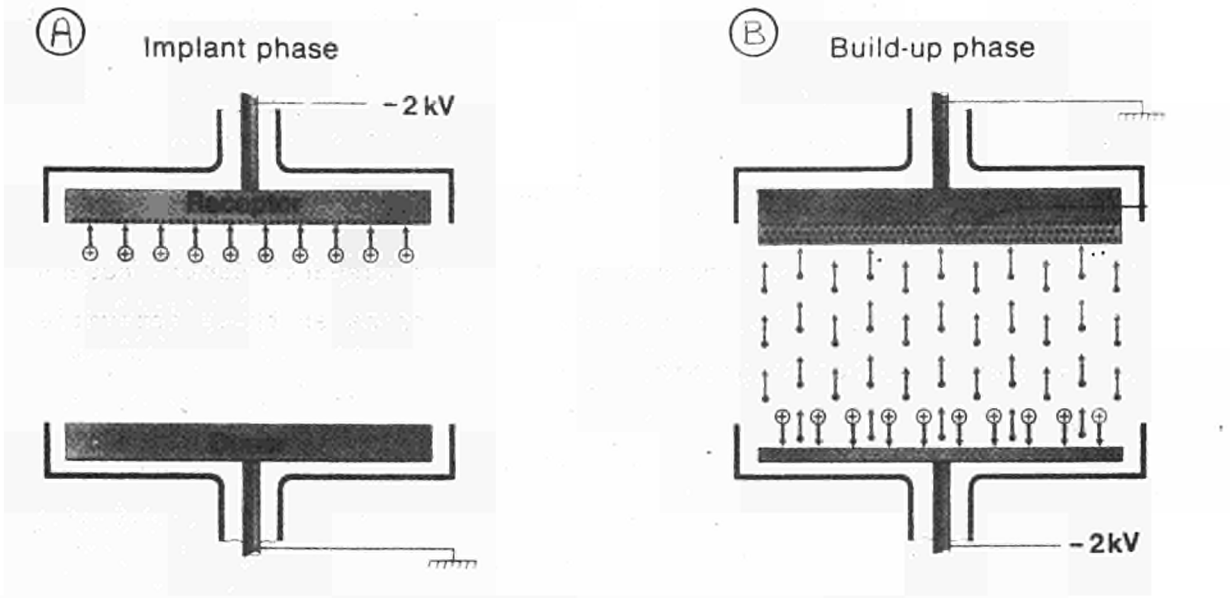


Fig. 8.2.1 : A - Implantation phase of Kr^+ ions into the metal
B - Coating phase by sputtering metal ions (reversed polarity)

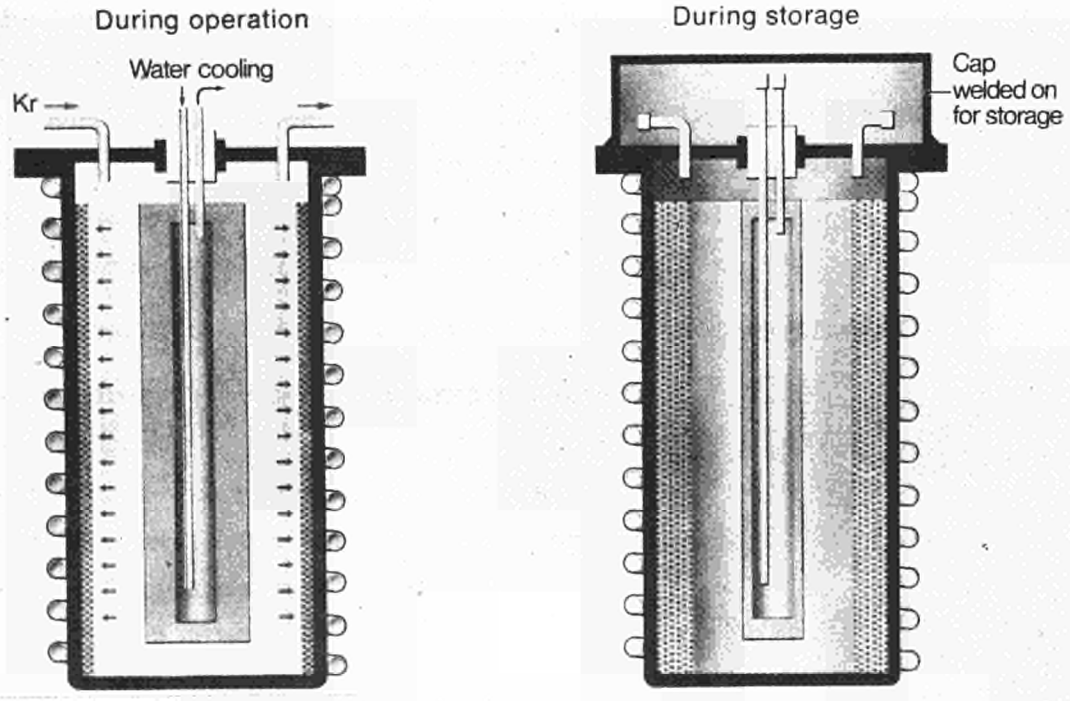


Fig. 8.2.2 : Plant unit

9. SEPARATION AND TRANSMUTATION OF ACTINIDES (UKAEA, HARWELL AND ECN PETTEN)

UKAEA and ECN have accomplished an assessment of the various techniques and strategies of actinides separation and nuclear incineration. The results of this study have been published (EUR 5801 e).

As the research work in progress at the Joint Research Centre covers most parts of this field, it was decided to transfer these activities to the direct action programme.

10. Review of problems raised by radioactive waste management which could not be solved within the framework of the existing legal, administrative and financial provisions

A study has been conducted by various national experts working under a contract with Environmental Resources Ltd, to determine the status of law applicable to radioactive waste management in the Member States of the Community. The final document consists of the various national reports and a consolidated report*.

The composition and main conclusions of the consolidated report are as follows :

Introduction : - origin of radioactive wastes, classification, quantities;
- practices followed in waste management and in legislation in the various Member States;
- competent national authorities.

It emerges that there is no legal definition of the term "radioactive waste", except in Germany and the United Kingdom.

The German definition explains the concepts "waste" and "radioactive", but does not establish a threshold below which an item of waste is no longer to be considered radioactive. The British definition only explains the term "radioactive" with figures to indicate the thresholds. The report considers that the German definition is, from the legal point of view, a good model for subsequent developments at international level.

The report then adopts a form of presentation comprising :

- temporary storage (recoverable wastes)
- transport
- disposal (non-recoverable wastes)
- discharge to the atmosphere and/or into water
- burial or immersion

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* Now being published; author : Professor E. de Pontavice (France)

This presentation would appear to form a basis for subsequent legal developments. It may be noted that it differs from the approach adopted by those responsible at the technical level for waste mananagement, which makes a distinction at the outset between the concepts of "dilution + dispersion" and "concentration + containment".

On the subject of legislative practice, the available data are very general and do not specifically concern radioactive waste.

Part I : Prevention of damage caused by radioactive waste

Intermediate storage seems to be adequately covered, as far as the radiation protection aspects are concerned, by existing national legislation based on the ICRP recommendations. Projects undertaken also seem to be adequately covered by existing legal procedures authorizing the construction of storage plants and/or the storage of wastes.

Transport of radioactive wastes appears to be adequately covered by the IAEA regulations on the transport of radioactive products and the corresponding legislation at national level.

The disposal of radioactive wastes (for the purposes of the report) is covered :

- with regard to the discharge of gaseous wastes to the atmosphere, by national legislation based on the ICRP recommendations;
- with regard to the discharge of liquid wastes into national waters, by the same legislation; discharge into international waters appears to be covered by various international conventions relating to carcinogenic substances in general;
- with regard to the underwater disposal of immobilized solid wastes, by the London Convention and the control procedure drawn up by the OECD Nuclear Energy Agency.

Disposal by burial underground is contemplated at present only by German legislation.

Part II : Civil liability for damage caused by radioactive wastes

The liability referred to seems to be covered, as far as temporary storage and transport are concerned, by national legislation based on the Paris and Brussels Conventions relating to nuclear operations in general. These Conventions have been signed by all the Member States of the Community, but the Netherlands and Ireland have not yet ratified them.

The report notes however the diversity of specific arrangements adopted by each country (amounts, time limits).

Civil liability with regard to final disposal of radioactive wastes is also governed, according to the authors of the national reports, by the above-mentioned rules applicable to temporary storage.

The report considers that this is indeed the case with the discharge of waste into water or to the atmosphere but expresses doubts regarding long-term storage and the disposal of wastes underwater.

It notes that, while the Paris Convention covers disposal sites, its application would involve liability on the part of the operator for an unlimited period, for the extinction period (10 years) takes effect from the time of the accident and not from the time of disposal (Article 8(b) of the Convention). It also points out that, in the event of an accident caused by the disposal underwater of wastes of different origin, the victims would probably have no recourse, for the Convention would require the operators responsible to be identified, which would probably be technically impossible. The report concludes that compensation for damage arising from the disposal of radioactive wastes is not fully guaranteed by the Paris Convention.

Third part : General provisions

All the national legislative arrangements include such provisions (fines, imprisonment). The report considers,however, that these provisions are much too weak in relation to the risks involved.

The report also considers that international law as it now stands provides no scope for the organization of an international system of control.

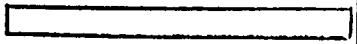
Conclusion : The routine management of radioactive wastes is covered in the Community as a whole by a licensing system which makes it possible to provide in what the report states is a fairly satisfactory manner for the protection of workers and the public, especially in view of the fact that these authorizations are based on international standards, which ensures a certain degree of uniformity in legislation.
The report also makes recommandations regarding the role to be played by the Community on this question.

11. LIST OF CONTRACTS

(indirect action)

INDIRECT ACTION

Programme sheet Nr. 1 : Immobilisation of medium active solide waste with plastic resins

Contractor	Project and major programme items	Est.tot. expenditure (MUA)	EEC Contribution (MUA)	Time scale 1976 1977 1978 1979	Contract N°
STEAG (D)	Immobilisation of medium active solid waste with plastic resins	0.48	0.12		027-76-7 WASD
SENA (F/B)	Construction of a pilot plant for immobilisation of medium active solid waste with plastic resins	1.67	0.36		039-77-6 WASFB
	Total	2.15	0.48		

Programme sheet nr. 2 - High activity solid waste : decontamination and conditioning of irradiated fuel element claddings

Contractor	Project and major programme items	Est.tot. expenditure (MUA)	EEC Contribution (MUA)	Time scale 1976 1977 1978 1979	Contract N°
CEN (B)	Press compaction and embedding in low melting alloy - Development of the process - Conceptual study of reference process	0.45	0.20	inactive simulated wastes Hot cell	030-76-11 WASB
CEA (F)	Decontamination by glass (Zry eutectic) and conditioning : - Laboratory experiment - Experiments at semi-industrial scale	0.39	0.17	simulated wastes	033-77-8 WASF
KFK (D)	Roll flattening and embedding in concrete : - Development of the process - Conceptual study of reference process - Comparative study of all methods	0.40	0.18	Const.+ Operate pilot(inact)	035-76-11 WASD
UKAEA	Characterization of radioactivities in cladding wastes : - Actinides : total content, surface and depth distribution - Fission & Activation products : total content, depth distribution	0.16	0.07	At least 8 batchs from : PWR, BWR, FBR, SGHWR, AGR (leached and P.I.E. samples)	037-76-11 WASUK
	Total	1.40	0.62		

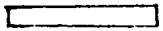
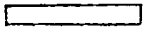
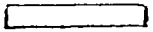
Programme sheet Nr. 4 : Treatment of combustible alpha wastes

Contractor	Project and major programme items	Est.tot. expenditure (MUA)	EEC Contribution (MUA)	Time scale	Contract N°
CEN/SCK (B)	High temperature incineration - Experiments with simulated alpha-wastes	1.17	0.50	1976 1977 1978 1979 80 inactive operation | active (β,γ) | α-active construc. | operate	017.76.7 WASB
UKAEA (GB)	Critical factors of Pu-recovery from ashes - Exploratory phase - Systematic experimental study	0.55	0.25		049-77-11 WASUK
UKAEA (GB)	Engineering study of pyrolysis process - Pyrolysis reactor (inactive) - Engineered off-gas, char handling systems	0.60	0.24	Study | Const. | operate	050-77-11 WASUK
AGIP NUCLEARE (I)	Molten salt Incinerator - Inactive lab. tests - Hot bench scale experiment - Inactive pilot plant	0.97	0.38	const. | operate construct. operate	036-76-7 WASI
KFK (D)	Acid digestion process - Digestion test facility (inactive) - Acid recycling & off-gas systems - Pu recovery test rig (alpha-active)	0.86	0.34	Construct operate site prep & construction oper.	044-77-7 WASD
GRAVATOM (GB)	Radiation assisted pyrolysis study - Preliminary feasibility study(continuation pending on results of this study) - Construction and Operation of experimental plant	0.27	0.11	Feasib.study constr. Oper.	045-77-11 WASUK
	Total	4.42	1.82		

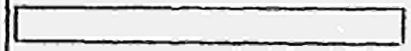
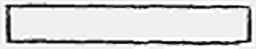
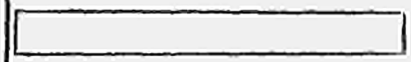
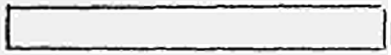
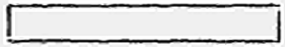
Programme sheet Nr. 5 : Joint programme of testing 5 reference substances of immobilization materials for solid HLW

Contractor	Project and major programme items	Est.tot. expenditure (MUA)	EEC Contribution (MUA)	Time scale 1976 1977 1978 1979 80	Contract N°
CEA (F)	Leach tests on active vitrified HLW blocks	0.18	0.08		040-77-11 WASF
UKAEA (GB)	Leach tests at various temp. & leach liquors	0.05	0.02	100°C | various T	034-77-1 WASUK
UKAEA (GB)	Divitrification versus leach rate	0.13	0.06	annealing | leaching	"
UKAEA	Alpha-radiation versus leach rate, helium release & Wigner energy	0.23	0.11	manuf. | irradiation | tests	"
HMI (D)	Divitrification effects on structure	0.25	0.10	instr. prep. | testing	029-77-1 WASD
	Total	0.84	0.37		

Programme sheet Nr. 6 : Joint study of air & water cooled interim storage for solid HLW

Contractor	Project and major programme items	Est.tot. expenditure (MUA)	EEC Contribution (MUA)	Time scale 1976 1977 1978 1979 80	Contract N°
BN (B)	Definition of basic schemes	0.10	0.05		041-77-10 WASB
NUKEM (D)	Conceptual design	0.13	0.07		042-77-7 WASD
NUKEM (D)	Risk analysis	0.07	0.03		042-77-7 WASD
BN (B)	Economic analysis	0.08	0.04		041-77-10 WASB
	Total	0.38	0.19		

Programme Sheet Nr 7 : Underground disposal

Contractor	Project and major programme items	Est.tot. expenditure (MUA)	EEC Contribution (MUA)	Time scale 1976 1977 1978	
GFK (D)	Disposal of radioactive waste in salt formations	2.81	1.34		015-76-1 WASD
ECN (NL)	Disposal of radioactive waste in salt formations	0.25	0.10		026-76-7 WASNL
CEN/ SCK (B)	Disposal of radioactive waste in deep clay formations	0.95	0.48		013-76-1 WASB
CNEN (I)	Disposal of radioactive waste in clay formations	1.40	0.70		020-77-9 WASI
CEA (F)	Disposal of radioactive waste in hard rock formations	1.60	0.70		019-76-7 WASF
DKAEA GB)	Disposal of radioactive waste in hard rock formations	1.44	0.54		018-76-7 WASUK
	Total	8.45	3.86		

INDIRECT ACTION

Programme sheet Nr. 8 : Storage of gaseous wastes

Contractor	Project and major programme items	Est. tot. expenditure (MUA)	EEC contribution (MUA)	Time scale 1976 1977 1978 1979	Contract Nr
ECN (NL)	Feasibility of Krypton sea disposal	0.08	0.03		052-78-6 WAS N
KFA (D)	- Theoretical analysis of problems related to Krypton sea disposal	0.09	0.03		051-78-1 WAS D
UKAEA	Tritium immobilization in a solid material : - Theor. screening study on candidate material	0.1	0.04		046-77-7 WASP
n.d.	Experimental study on tritium immobilization (Work to be defined after preliminary assessment)	~ 0.15	0.06		
CEN (B)	Incorporation of iodine in plastic resins : - Inactive experimental study - Leaching test (trace of I-129)	~ 0.12	0.04	12 months	
n.d.	Krypton encapsulation in zeolites : - Experimental study of the method	0.17	0.06		

Programme sheet Nr. 8 : Storage of gaseous wastes

Contractor	Project and major programme items	Est.tot Expenditure (MUA)	EEC Contribution (MUA)	Time scale 1976 1977 1978 1979	Contract N°
LE VERRE FLUORE s.a.r.l. (F)	Incorporation of iodine in low melting glass : - Laboratory experiments	0.25	0.09		054-78-1 WAS F
CEN (B)	Charcoal absorbed Krypton storage in cylinders (low pres.) : - Experimental study	0.14	0.06		048-77-1 WASB
UKAEA (UK)	Krypton incorporation in a metallic matrix : - Design, construction and commissioning of a pilot plant (half-scale) - Pilot plant operation			active	043-77-7 WASUK
KFA (D)	Krypton storage in pressurized cylinders : - Laboratory experiment - Conceptual study of engineered storage unity	0.17	0.08		047-77-7 WASD
	Total	1.84	0.76		

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13. MEMBERS OF THE ADVISORY COMMITTEE FOR PROGRAMME MANAGEMENT "MANAGEMENT AND STORAGE OF RADIOACTIVE WASTE" (AT THE 31 DECEMBER 1977)

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14. LIST OF ABBREVIATIONS

AEC-RISØ	Atomenergikommissionens Forsoegsanlaeg Risó (Danmark)
BGR	Bundesanstalt für Gewissenschaften und Rohstoffe
BN	Belgonucléaire - Bruxelles
BRGM	Bureau de Recherches géologiques et minières
CEA	Commissariat à l'Energie Atomique - Paris
CEN/SCK	Centre d'Etudes de l'Energie Nucléaire/ Studiecentrum voor Kernenergie Mol - Belgique
CNEN	Comitato Nazionale Energia Nucleare - Roma
ECN	Energieonderzoek Centrum Nederland - Petten
GRAVATOM	Gravatom Industries Ltd - Gosport (UK)
HMI	Hahn Meitner Institut - Berlin
KFA	Kernforschungsanlage Jülich GmbH - Jülich
KFK	Kernforschungszentrum Karlsruhe GmbH - Karlsruhe
NEB	Nuclear Energy Board (Ireland)
NERC	National Environment Research Council
NUKEM	Nuclear-Chemie und Metallurgie GmbH, Wolfgang/Hanau
SENA	Société d'Energie Nucléaire Franco-Belge des Ardennes - Chooz (Ardennes)
STEAG	Steag Kernenergie GmbH - Essen
UKAEA	United Kingdom Atomic Energy Authority

European Communities — Commission

EUR 6128 — The Community's R & D Programme on Radioactive waste management and storage (second annual progress report)

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The European Community's programme is the first and to this date the only joint international action dealing with those issues, which might well become decisive for the future of nuclear energy – the management and storage of radio waste. The present Annual Progress Report describes the scope and the state of advancement of this indirect action programme up to 31 December 1977 and follows the 1976 Annual Report (EUR 5749). At present about 50 research contracts with research institutes in almost every member country of the EC are either signed or in the final stages of negotiation.

The objective of the R & D actions to be achieved by 1980 is the demonstration of either the technical potential or, for further advanced projects, the feasibility and even the industrial availability of methods for treating and storing radwaste. The following aspects are investigated:

— processing of solid waste from reactors, reprocessing plants and the plutonium manufacture;
— intermediate and terminal storage of high activity and alpha wastes;
— advanced waste management methods as the storage of gaseous waste.

In addition to the scientific-technical R & D actions, a survey of the legal, administrative and financial problems encountered in radwaste management and storage is an essential part of the Communities' programme.

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