COMMISSION OF THE EUROPEAN COMMUNITIES

nuclear science and technology

The Communities R & D Programme

RADIOACTIVE WASTE MANAGEMENT AND STORAGE

(First annual Progress Report)

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Directorate-General
1977 Research, Science and Education, Brussels (Belgium) EUR 5749 e, f

Abstract

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 radioactive waste.
The first Annual Progress Report describes the scope and the state of advancement of this indirect action programme. At present 24 research contracts with research institutes in almost every member country of the EC are either signed or in the final stages of negociation.

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 stoping 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 and the separation and transmutation of actinides.

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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FOREWORD

This is the first progress report concerning the programme on radioactive waste management and storage (indirect action) of the Commission of the European Communities.

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.

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.

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* O.J. Nr. L 178/28, 9.7.1975

** 19,16 Mio u.a. = approx. 1,30 US $ on 1.1.77.

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 structures ;
- 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 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).

This report covers the period from 1st January 1976 to 1st December 1976.

The next progress reports will be issued on a yearly basis.

1. TREATMENT OF LOW AND MEDIUM LEVEL WASTE

1.1 Mobile Pilot Plants for Resin Imbedding of Reactor Waste

1.1.1 Objective and scope of the programme

The contract with STEAG Kernenergie GmbH aims at the development of a technique to immobilize and condition low and medium active reactor waste (mainly from water reactors).
The intention is to imbed the waste in plastic resins. The resulting solid products shall be examined in respect to their compliance to the requirements for intermediate and terminal storage. Active operation of a pilot plant should demonstrate the industrial availability and range of application of the techniques developed under this contract.
The programme will proceed in five stages :

a) Preliminary investigation
b) Study and choice of candidate resins and conditioning methods
c) Laboratory studies and technical experiments with inactive and active waste
d) Commissioning tests and experimental operation of the pilot plant on a reactor site
e) Data processing and analysis of the results.

1.1.2 Preliminary investigation

1.1.2.1 Range of application

The various categories of reactor waste (see Fig. 1) should be reviewed in order to determine, for which types of wastes the proposed technique promises to be applicable.
The common waste types should be grouped according to the urgency of treatment in respect to the amounts and volumes generated, the risk arising to the public and the lack of satisfactory techniques of immobilization.

1.1.2.2 Requirements concerning the product (conditioned waste)

A set of preliminary requirements and conditions of acceptance of the product for storage shall be drawn up.

The main criteria for acceptance of this type of solidified waste to storage facilities will be :

- leach resistance,
- radiation resistance,
- structural strength,
- long term physical and chemical stability,
- resistance against bacteria and
- flammability.

1.1.2.3 Requirements concerning the resin

The criteria for the choice of the resins will also be examined. These will be defined to comply with the following requirements :

- safety of handling (flammability, toxicity of the resin)
- safety of storage and in the process,
- ease of control of the chemical reactions and
- compatibility with the incorporated waste.

1.1.3 Study of the resins and the conditioning techniques

1.1.3.1 Choice of suitable resins

The criteria established in 1.1.2.2 and 1.1.2.3 permit a first selection of candidate materials. Although information on most of the relevant properties is available, some data may have to be measured or verified by lab experiment.

The following general categories of resins have been short listed :

a) polyurethanes ,
b) unsaturated polyesters,
c) polystyrenes and
d) epoxy resins .

Further alternatives can be examined, if promising.

1.1.3.2 Testing of the candidate materials

The following aspects will be investigated by laboratory experiments :

- chemical compatibility of the resin with (simulated) waste ,

- variation of the reaction rate with waste composition,
- determination of leaching rate,
- influence of additives for controlling the solidification and water absorbtion,
- optimal ratio of waste/resin volumes.

1.1.3.3 Technical experiments

The best mixing technique for obtaining a homogeneous product will be determined. The unit operations to be tested are :

- pouring,
- injecting,
- sedimentation and
- stirring.

For this purpose pilot-scale experimental equipment will be assembled.

1.1.3.4 Prototype development

Once the suitable mixing operation has been determined, further technical experiments on existing equipment are scheduled with inactive, simulated waste.

These tests are intended to provide the necessary information :

- on the mixing stage,
- waste preparation (drying, chemical pre-treatment),
- control of solidification reaction and reaction heat,
- off-gas treatment (if necessary),
- transport of moist waste from the tanks to the conditioning plant,
- remote control mechanisms and
- corrosion and wear of moving components.

A series of tests simulating certain accidental conditions will be run, if these conditions cannot be assessed by other means.

The operating conditions and the production sequence will be examined under the aspects of safety, reliability and economy.

Further tests for the development of the main plant components will use representative, low-active waste. These tests serve to assess :

- the operational radiation exposure to the personnel ,
- the contamination and the radiation hot spots of the plant.

Finally these work tests will provide representative specimen of the product, which will be tested against the criteria under 1.1.2.2, in particular :

- homogeneity of waste-resin mix,
- leach resistance,
- volume ratio waste/resin (or waste/product) obtainable,
- safety and economy of packing material and
- radiation stability of the product.

1.1.4 Operation of a prototype plant

The design and construction of the prototype plant are not covered by this contract, as STEAG has undertaken to build the plant for commercial exploitation and finances this investment itself.
However, the commissioning phase and a first series of operation runs of the mobile plant on a reactor site that will provide the final experimental evidence for the validity of the technical concept, are incorporated in the Community programme.
All operative data and the experiences gained until 31 December 1979 will be reported.
Samples of routine and non-routine runs will be examined and the reliability and availability of the plant will be recorded.
The radiation exposure of the personnel will be reported by the station's health physicists.
The licences for the operation of the plant on a reactor site and for acceptance of the conditioned product for transport and terminal storage at the Asse salt mine will be obtained by STEAG.

1.2 Stationary resin imbedding with the CEA technique

Contractual negociation towards participating in the operational experience of a polyester resin imbedding plant at Chooz will only begin in 1977.

2. DECONTAMINATION AND CONDITIONING OF IRRADIATED FUEL ELEMENT CLADDING

2.0 Introduction and description of waste

The claddings of fuels irradiated in nuclear reactor have after removal of the fuel still a high radioactivity, which identifies them as high level waste. There are two sources of this activity :

- activation products formed by neutron exposure of constituents of the cladding or deposited by reactor coolant
- fission products, transuranic elements from the fuel and their interaction products. These may be present as undissolved fuel, as a surface layer or inside the cladding material, as a result of diffusion or implantation.

The waste arising from the reprocessing of fuels, contains, in addition to hulls, structural materials such as spacers, springs, tie rods and end pieces.

The present expedient of under-water storage in concrete silos on the premises of the reprocessing plant is not a satisfactory long-term solution.
The research programmes investigating the various decontaminating and conditioning methods and characterizing the activity of the hulls are described in Chapter 2.1 to 2.4.

2.1 Incorporation in low melting alloys (CEN)

The objective of this study is to develop a method of conditioning of cladding waste, which consists of press compaction and embedding of the hulls in a low melting alloy. The final product had to be appropriate for long-term storage.

The reference material is the cladding waste generated by reprocessing of light-water-reactor fuels. This metal scrap, having a density of about 1.1 Kg/dm3, is composed of Zircaloy-4 hulls (70-80 wt %) and structural materials in inconel and stainless steel. The research works, already started on 1st November 1976, will be accomplished at the end of 1979.

2.1.1 Study of filling alloys

2.1.1.1 Literature review and basic selection of filling alloys

Literature data on the following characteristics of potential filling alloys will be compiled : melting point, thermal expansion, contraction at solidification, interactions with cladding waste materials, corrosion in water, cost and availability.
On the basis of these data suitable alloys will be selected for investigation in the present study.

2.1.1.2 Experimental investigation of suitable alloys (inactive tests)

In case of insufficient literature data the following phenomena will be studied :

- Interactions with waste components (Zircaloy, stainless steel, Inconel),
- Wettability of waste components
- Corrosion of samples with and without embedded waste components in pool water and ground water.

The following analytical techniques will be used :

- microscopic examination and / or microprobe analysis
- measurement of weight change for corrosion experiments
- chemical analysis.

2.1.1.3 Hot cell tests with cladding samples from spent fuel elements

a) Tests and analytical techniques
   - Dispersion of transuranics and fission products in filling alloys ; alpha and gamma radiography
   - Long-term leach tests on cladding samples embedded in filling alloy ; gross alpha and gross beta-gamma radiometry ; alpha and gamma spectrometry

b) Cladding samples
   - Origin : BR-3 fuel elements irradiated to a burn-up of 15 to 30 MWd/kg
   - Condition : Discutting of samples, dissolution of fuel in nitric acid followed by washing with nitric and water.

2.1.2 Experimental investigation of press compaction

Preliminary tests will be done in order to establish that satisfactory penetration of compacted blocks can be achieved. If the result of these tests is positive, as is expected, work will be continued as outlined in paragraphs 2.1.2 and 2.1.3, and work outlined in paragraph 2.1.4 will not be done. If the result of the preliminary tests is negative, work is negative, work will proceed on paragraph 2.1.4 instead of paragraphs 2.1.2 and 2.1.3, which will be dropped.

a) Results to be obtained
- Determination of required press force
- Effects of loading height
- Effects of grids and massive structural materials on densification factor
- Measurement of heat generated during compaction

b) Operating conditions
- Inactive material without and with pretreatment (hydrogen embrittlement in order to simulate post irradiation conditions)
- Presses of 150 t and 600 t force
- Maximum diameter of die : 150 mm
- Atmosphere : air
- Temperature : ambient

c) Examination

Determination of density and porosity.

2.1.3 Development of a process for filling compacted blocks

a) Results to be obtained
- Development of the appropriate procedure
- Optimizing of process parameters (filling temperature ; pressure conditions ; etc)
- Quality control of product (completeness of filling, wetting)

b) Operating conditions
- Inactive material
- Engineered test unit (to be constructed)
- Maximum diameter of briquette : 150 mm

c) Examination

- Metallography
- Determination of porosity.

2.1.4 Development of a process for compaction in presence of filling alloy

(This process promises better assurance of complete filling than the process considered in paragraph 2.1.3)

a) Results to be obtained

- Development of the appropriate procedure
- Optimizing of process parameters (pressing temperature, etc)
- Quality control of product (homogeneity of filling, wetting)
- Other results as in 2.1.2.a)

b) Operating conditions

- Inactive material
- Engineered test unit (to be constructed)
- Presses of 150 t and 600 t force
- Maximum diameter of die : 150 mm

c) Examination

- Metallography
- Determination of porosity

2.1.5 Conceptual study of a reference process

This study will provide the information needed for the comparison of the different methods. Requirements for a 300 tU/y reprocessing plant will be evaluated and the results will also be extrapolated to a 1500 tU/y plant. Cost estimates for both plant capacities will be provided.

2.1.5.1 Definition of a reference process

Selection of filling alloy, process variant (filling of compacted blocks or compaction in presence of filling alloy) and main process parameters.

2.1.5.2 Definition of optimal dimensions and of packaging of the briquettes

This will include :

- Heat transfer calculations
- Evaluation of shielding requirements
- Consideration of manipulation aspects

2.1.5.3 Safety and environmental impact

The following aspects concerning the safety and environmental impact will be studied :

- Evaluation of process hazards, particularly due to pyrophorocity of Zircaloy ; definition of the required engineered safeguards.
- Estimation of secondary waste and effluents produced.

2.1.6 Conceptual study of a hot cell unit

This experimental unit shall be able to perform all operations of the reference process with the active waste material.
The study will include :

- Consideration of safety aspects
- Conception of the remotely operated machinery and supply of assembly drawings
- Cost estimate.

This unit shall provide at a later stage the complementary information required for the industrial application of the reference process.

2.2 Incorporation in concrete (G.f.K)

The research programme of the contract is aimed at the following topics :

- Development of a special conditioning method :
  Volume reduction by rolling and subsequent embedding in concrete.
- Theoretical comparative study of the different conditioning methods.

The reference material is the same as defined in chapter 2.1. All conceptual studies and cost estimates will be based on an amount of 580 t. waste per year, arising from a reprocessing plant with a capacity of 1400 t U/year.
The programme will be carried out from 1st November 1976 to the end of 1979.

2.2.1 Conditioning by rolling and embedding in concrete

2.2.1.1 Characteristics of concrete

A review of candidate concrete types and the criteria of selection will be given.
Furthermore the most relevant concrete properties required for a successful conditioning will be described.

2.2.1.2 Development of the process

Preliminary tests with inactive materials on deformability by cold rolling will determine the required press force and also clarify the following points :

- Obtainable compression ratio (volume reduction factor)
- Influence of inconel and stainless steel structural pieces.
- Cracking of the material.

Conditioned samples at various stages of compaction will be produced with inactive and active material. These samples, which are later needed for the tests described in chapter 2.2.1.4, will also provide information on the deformability of irradiation damaged materials and on the quality of the concrete filling.

The active samples should be taken from light-water reactor fuel elements with a high burn-up (20.000 + 30.000 MWD/MTM). After cutting and dissolution of fuel, the leached out hulls will be washed with nitric acid.
Some pieces of zircaloy hulls, dissolved in acid, will be submitted to ray-spectrometry for identification of the radionuclides. These results are needed to determine the leaching rate of the conditioned final product.

2.2.1.3 Testing of the process

It is planned to build an inactive pilot plant, working in inert atmosphere, for testing and optimizing the techniques of conditioning. This installation will include : a storage vault, the feeding equipment, the compaction roller, the container filling station, the concrete dosing and the container shaking equipment.

2.2.1.4 Examination of conditioned wastes for disposal

The most important characteristics for the evaluation and the choice of a conditioning technique will be measured as follows :

- Measurements of heat conduction as function of compaction and temperature will be carried out on inactive samples.

Conditioned waste samples of different densification will be irradiated. Afterwards irradiated and unirradiated samples will be submitted to the following examinations :

- Determination at room-temperature as well as at elevated temperature of the physical and mechanical properties such as : heat conduction, combustibility, crushing resistance, etc.
- Laboratory determination of free and retained gases, including tritium, evaluation of radiolysis and the gettering effect of zircaloy.

The leaching resistance of active conditioned samples will be measured in distilled water and in salt solution (on the basis of the IAEA recommendation reported by "Atomic Energy Review" 9 (1971), 195).

2.2.1.5 Conceptual study of a reference process

The optimized parameters for a large scale application of the developed method will be established from the experimental results. The following parameters and aspects will be studied :

a) Dimension and packing of conditioned wastes.
They will depend upon :
- the calculated central temperature and maximum admissible value assessed for a final deposit,
- required shielding,
- handling aspects.

b) Safety
A safety analysis of the process as a whole will be carried out ; in particular the risk of spontaneous zircaloy combustion and the release of gases will be taken into account.
The amounts and activity of the secondary wastes produced by the process will be estimated.

c) Cost estimate
An assessment of costs will provide the data for the choice of the most economical solution.

d) Conceptual design of a large scale plant
A design study for the industrial application of the reference method including the auxiliary equipment will be produced. The study will incorporate general assembly drawings.

2.2.2 Intercomparative study of different methods

2.2.2.1 Methods

The various candidate processes for treating cladding wastes will be compared, in particular the following methods developed under the EEC waste programme :

- Densification by rolling and embedding in concrete;
- Press compaction and embedding in low melting alloys;
- Decontamination of molten wastes (eutectic) by glass and conditioning of the products ;
- Overcoating with glass.

2.2.2.2 Aspects of evaluation

The study has to consider all the aspects which could determine the choice of the process for the industrial application and particularly ;

- Risks of conditioning, transport and intermediate storage ;
- Long term hazards of an appropriate ultimate disposal ;
- Costs ;
- Secondary wastes produced
- Possibility of zirconium recovery.

2.3 Decontamination and conditioning in glass (CEA)

The aim of this study is to devise a method of decontamination and conditioning for cans in a form which will permit long-term storage.

Two different processes have been considered :

- Compaction by eutectic fusion and high-temperature decontamination by the introduction of glass into the metal bath.
  Separation of slag and final conditioning of the slag and of the alloy.
- Imbedding in glass, used elsewhere for the solidification of fission products.
  Most of the work envisaged, and in particular the whole of the laboratory study, involves the first process.

The materials for study will be those obtained from the decladding of fuel irradiated in light water reactors, breeder reactors and possibly reactors using fuels with the same type of cladding.

There are two main groups :

- Zircaloy alloys, particularly Zircaloy 4 (reference ASTM : Standard B-353 grade RA2)
- Stainless steels of similar composition to alloy AFNOR Z 2 CND 17/12 (reference AISI : 316 L; reference Aubert and Duval : X 18 M Z W).

The quantity of stainless-steel claddings is small compared with that of Zircaloy claddings.

The materials will be in the form of tubes varying in length from 10 to 20 mm, of outside and inside diameters in the order of 7 and 6 mm respectively. These tubes are radioactive activates and are contaminated by heavy radioactive elements and possibly by fission products.

It may be assumed that summary decontamination by acid leaching has been carried out in the reprocessing plant.

The study will be based on the treatment of claddings arising from the reprocessing of 1 500 tonnes of irradiated fuel per year which envolves approximately 400 tonnes of hulls per year.

The programme will begin on 1 March 1977 and will be completed on 31 March 1979.

2.3.1 Laboratory study

2.3.1.1 Eutectic mixture

A series of experiments will be carried out to develop eutectic mixtures of zirconium and stainless steel with one or more other metals or metalloids aimed at lowering the temperature required for fusion.

The tests will be conducted in an electric furnace equipped, if necessary, with a sealed chamber to provide a controlled atmosphere on quantities in the order of several hundred grammes. The furnace hearth will be made of siliceous alumina materials or of sintered alumina (Desmarquest type). The atmosphere used will be argon.

The following filler metals will be used to begin with :

- Iron (content in the region of 16 %)
- Copper (content in the region of 20 %)
- Stainless steel, types Z_3 CN 18-10 and Z_2 CN 18-12 (contents of 15 % to 20 %)
- Nickel alloy, Inconel 601 type
- Carbon, silicon, boron.

The following results will be established :

- Lowering of fusion point by the use of a particular filler metal or metals.
- Determination of the optimum content of these metals in the alloy.
- Influence of the atmosphere used during fusion on the formation of the eutectic.
- Evaluation of the speed at which the Zirconium and stainless steel are absorbed.

2.3.1.2 Refractory materials

The selection of the refractory material or materials most appropriate for the fusion of eutectic and stainless steels will be determined by a corrosion study.

The service life will be evaluated in terms of the nature of the alloy, the atmosphere and the temperature.

The following materials will be studied in particular :

- Inker Dl (calcined magnesite)
- CaF_2
- Graphite
- ZAC 1681 and ZAC 1711 (Al_2O_3 + SiO_2 + ZrO_2; melted)
- Magnalux (Al_2O_3 + SiO_2 mainly; melted)
- SiC
- Cr_2O_3
- Sintered magnesia.

The sample to be tested will be immersed in the liquid mixture (metal and glass) for 100 hours at a temperature 50-100° C in excess of that of the liquidus in an argon atmosphere. The tests will be carried out either with the zirconium eutectic or with stainless steel in the molten state. The container used will be made of zirconium.

Corrosion will be evaluated by visual and/or microscopic examinations, by photographs and the weighing of samples.

2.3.1.3 Decontaminating glass and characterization of the final products

Borosilicate, aluminoborate and phosphate glasses will be the subject of laboratory experiments to produce the following results :

- Influence of the composition of the glass, with particular reference to the reaction with zirconium
- Evaluation of decontamination for uranium and fission products.
- Optimum ratio of glass to metal.

The operating conditions envisaged are as follows : addition of glass after melting of the alloy, addition of products during feed and separation of slag.

The conditioned products will be characterized for the purpose of long-term storage.

The following examinations will be carried out : chemical analysis of slag, determination of physical properties (melting point, viscosity) and crystallography.

2.3.2 Technological study

2.3.2.1 Preliminary tests on the design of prototype plants

Various simplified fusion rigs for discontinuous or continuous operation will be devised and tested in order to facilitate the definition of furnace characteristics required.

Initially, the melting process will be carried out in a tilting electric induction furnace, supplied with power by an inverter having the following characteristics :

- Nominal frequencies : 3 000 Hz and 10 000 Hz
- Nominal output at mean frequency : 50 kW.

These plants will be completed once suitable techniques have been selected for the construction of plant for use in experiments conducted without activity.

2.3.2.2 Ancillary devices

Regarding the first process-compaction by fusion - devices will be studied for casting, separation of slag and conditioning. For the second process - incorporation in a glass matrix - a device will be developped to achieve the necessary homogeneous dispersion.

2.3.2.3 Experimentation with prototype plants

A special furnace will be constructed for the development of a prototype plant, which will be used to carry out tests with simulated claddings. These could be in the form of stainless steel and zirconium tubes with dimensions similar to those of the claddings, zirconium plates or, equally, rejected claddings.

Decontamination tests on this plant will be carried out with uranium-contaminated claddings. The decontamination will be evaluated by analytical determination of the uranium.

2.3.2.4 Design of the plant for experiments under radioactive conditions

The data obtained from the experiments involving the first process will be evaluated and design principles will be established for the construction of an active plant with a melting capacity of 50 kg per charge.

2.3.3 Study of a reference process

This study should supply the data making possible an initial comparison of the proposed process with the other processes studied under the Community programme.

The reference process will be defined by the choice of procedures and parameters appropriate for industrial application, including cost estimates.

2.3.3.1 Dimensions and packaging of conditioned products

After evaluation of the heat to be removed and the shielding required and after consideration of the handling aspects, the optimum dimensions and the type of packaging will be determined for the conditioned waste.

2.3.3.2 Safety

The safety aspects relating to the process will be examined with particular reference to the pyrophoricity of Zircaloy. Indications will be giving concerning the necessary preventive measures. The production of secondary waste and effluents will be estimated.

2.4 Characterization of the radioactivity in different cladding wastes (UKAEA)

The research programme is aimed at identifying the nature, location and activity levels of the radioactive components in typical irradiated fuel element claddings. The quantitative and qualitative information provided will be of assistance in any subsequent treatment.
The study will cover Zircaloy mainly and stainless steel coming from the operation of nuclear reactors as : PWR, BWR, SGHWR, AGR and FBR. Priority will be given to specimens of leached hulls arising from fuel reprocessing plants. Samples obtained from the post-irradiation examination (PIE), the position of which along the length of a fuel pin can be identified, will also be considered.

Table 1 gives a survey of the sample proposed for investigation. Already available specimens shall be examinated at first and further batches shall be selected according to availability and priority of interest. The whole programme will be carried out in the period from 1st November 1976 to the end of 1979.

Table 1 : List of candidate hulls samples for examination

Batch number	Cladding Material	Origine of samples Organism (installation)	Reactor type and/or name	Remarks
1	Zry	GfK/WAK (R.P.)	PWR, KWO	already available
2	SS	UKAEA (P.I.E.)	AGR	" "
3	Zry	UKAEA (P.I.E.)	SGHWR, Winfrith	" "
4	SS	UKAEA (L.S.R.)	FBR, Dounreay	" "
5	Zry	GfK/WAK (R.P.)	BWR, KRBI	not yet available
6	Zry	GfK (L.S.R.)	BWR-$(U,Pu)O_2$ fuel	" " "
7	Zry	UKAEA/BNFL (R.P.)	PWR	to be identified
8	Zry	UKAEA/BNFL (R.P.)	PWR	" " "
9	Zry	GfK (P.I.E.)	PWR, KWO	not yet available
10	Zry	CEA/COGEMA (R.P.)	LWR	recovery problems
11	SS	CEN (P.I.E.)	Rapsodie, Core Fortissimo	not yet available
12	Zry	UKAEA/BNFL (R.P.)	PWR	future reprocessing

Zry = Zircaloy ; SS = stainless steel ; R.P. = reprocessing plant ; P.I.E. = post-irradiation examination ; L.S.R. = Laboratory-scale reprocessing.

2.4.1 Visual inspection

Hulls will be visually inspected to observe defects, deposits and any anomalous phenomena which could be useful for the later interpretation of the analytical results.

2.4.2 Surface distribution of actinides

The actinide concentration per unit of surface and its distribution will be determined by alpha-spectrometry.
This will be performed on samples whose gamma activity is not too high for handling.
Actinide concentration would be estimated only if actinide composition is known or can be measured.

2.4.3 Total content of actinides and of fission and activation products

Different techniques and methods will be used as described in the paragraphs 2.4.3.1 to 2.4.3.3.

2.4.3.1 Neutron activation techniques

The delayed neutrons resulting from fission after irradiation in a thermal neutron flux will be measured. The counters will be standard gas filled neutron detectors (either tritium or BF_3). The method does not differentiate between different nuclides but the approximate total content of fissile nuclides will be obtained.

2.4.3.2 Gross alpha-count and alpha-spectrometry

Samples from individual hulls from each batch will be completely dissolved. At first the gross alpha-count on all sample solution will be carried out. Afterwards alpha-spectrometry will be performed on sample solutions, selected according the previously measured alpha activity, for the quantitative determination of :

- Pu-238, Pu-239 and Pu-240
- Am-241
- Cm-242 and Cm-244

In order to perform the above mentioned quantitative determinations, chemical separations of americium will be made. The alpha spectrometer consists of a gridded ionisation chamber coupled to a multi-channel analyser. Samples will normally be mounted directly from aqueous solution. Where appropriate the organic extractant phase will be mounted. The overall accuracy on the obtained mean values of results will be estimated. Where necessary the measured concentrations will be corrected for decay to the date of reactor shut-down. Alpha-spectra will be recorded in analogue and digital form.

2.4.3.3 Gamma-spectrometry

Sample solutions will be submitted to the determination by means of gamma-spectrometry, of the following fission and activiation products :

Sb-125, Cs-134, Ru-106, Cs-137, Ce/Pr-144, Mn-54, Co-60, Zr/Nb-95.

The gamma ray spectra will be measured at least on two parallel aliquots of each solution, using a (Ge-Li) detector.
Accuracy of results, decay correction and spectra recording form will be as mentioned in paragraph 2.4.3.2.

2.4.4 Depth distribution of actinides and fission products

The depth distribution of actinides and fission products will be investigated by different techniques.

2.4.4.1 Fission track analysis

Fission track autoradiography shows the distribution of fissile nuclides in the hull sections examinated. The polished specimen, coated with a plastic solid-state track detector is irradiated in a known thermal neutron flux in the DIDO reactor. The detector is then separated from the sample, treated chemically and examined microscopically to determine the damage caused by fission products. The results will be shown photographically and relative track densities will indicate the amounts of fissile material present.

2.4.4.2 Alpha-spectrometry on hulls

The depth distribution of actinides near the surface will be evaluated by means of alpha-spectra taken from the hulls themselves.

2.4.4.3 Alpha and gamma measurements

Where feasible, solutions will be prepared by chemical dissolution of successive layers of at least two hulls samples per batch.
These solutions will be examinated by alpha counting and gamma spectrometry in order to determine the concentration of actinides, fission and activation products.

2.5 Assessment of the various types of cladding

The Advisory Committee for Programme Management agreed that the Commission treats this subject by means of an inquiry. This inquiry has to provide a prospective inventory, until 1990, of cladding wastes composition and the expected amount arising in the European Community from spent fuel from the following reactor types : PWR, BWR, SGHWR, AGR, FBR.
A draft summary report (Doc. DG III/E-1, BHa/tg of 1st December, 1976), giving a survey of the answers to the inquiry, was prepared and sent to the experts of the Member States asking for suggestions or remarks. Missing or incomplete answers were completed with own estimates.

2.5.1 Cladding wastes

The information received on the amount of waste for each of the different reactor and cladding types is summarized in Table 1.
In case of diverging data for the same reactor and cladding type, the two extreme values are given.

Table 1: Cladding wastes from different reactor types

Reactor type	Cladding material (x) (percentage of total)	Total amount of waste in kg/ton H.M.
PWR	Zircaloy - 4 (70 w/o)	324 - 398
PWR	AISI - 304 (75 w/o)	350
BWR	Zircaloy - 2 (85 w/o)	293 - 400
BWR (Th-U fuel	AISI - 304 (65 w/o)	415
CANDU - BLW	Zircaloy - 4 (30 w/o)	370
SGHWR	Zircaloy - 2 (75 w/o)	282
AGR	Stainless steel (80 w/o)	154
FBR (Core)	AISI - 316 (40 w/o)	1422 - 1745
FBR (radial)	AISI - 316 (30 w/o)	849

(x) Including end plugs.

2 .5.2 Waste arisings from reprocessing

On the basis of the known time schedules for the planned reprocessing capacities in the Member States to come on stream, the cumulative expected amounts of cladding wastes were estimated. A survey of these "high" forecasts until 1990 is given in Table 2. The cumulative amount of wastes in the European Community will rise from about 4000 m. tons in 1985 to about 1600 m.tons in 1990. These values could be reduced by half in the case of the "low" estimates, which take into account possible delays in bringing new plants on stream or any other lowering factors of the actual reprocessing capacities.

Table 2 : "High" estimated cumulative arisings of cladding waste (tons)

Reprocessing plant	Fuel origin	Cladding Waste (m.tons) 1980	1985	1990
Cap de la Hague	LWR *	600	2550	6500
Windscale	AGR SGHWR LWR *	- - -	750 - 280	2260 725 1760
W.A.K.	LWR	65	115	115
KEWA		-	145	2650
MOL	LWR	-	175	660
ITREC - Rotondella	LWR	-	15	1300
Total in European Community		665	4.030	15.970

(*) estimated values

2.5.3 Radioactivity

The received data on radioactive isotopes formed by neutron activation of metal components were elaborated. Contaminating substances were not taken into consideration. The total activities of the materials, based on neutron exposure rates of $2 \cdot 10^{21}$ and $2.8 \cdot 10^{21}$ n/cm^2 (thermal flux), are given in Table 3 as function of decay times after reactor shut-down.

Table 3 : Total activities of waste materials (Ci/kg)

Material	Total activity (Ci/kg) after cooling for		
	1 y	3 y	10 y
Zircaloy	7 - 8	0.4 - 0.6	0.1
Inconel	40 - 180	25 - 120	22 - 55
Stainless steel	150 - 260	75 - 160	25 - 40
Harmonic steel	350	200	30

For Inconel and stainless steel the considerable differences in activity, which cannot be explained by the different neutron exposures, should be cleared. Variations on the composition of alloys can only partially give an explanation.

Stainless steel irradiated in FBR will reach, after an exposure rate of about $2 \cdot 10^{23}$ n/cm^2 and 1 year cooling time, a total activity of 120 Ci/kg.

3. INCINERATION OF PLUTONIUM CONTAMINATED WASTE

3.1 High Temperature Incineration (SCK/CEN)

3.1.1 Objectives of contract

Two strategies will be pursued in course of the programme :

a) Incineration of low Pu-contaminated waste with the aim to produce a highly leach resistant product suitable for long term storage and ultimate disposal.

b) Incineration of heavily Pu-contaminated waste with the aim to produce a readily soluble product in order to promote maximum Pu-recovery. The bulk of low contaminated waste will be processed in the existing FLK 60 furnace.

3.1.2 General description of incinerator

CEN's objective is to develop an incineration process which decomposes the most common Pu-contaminated waste materials by complete combustion in connection with a safe gas purification system. The flow sheets Figs. 3.1.1 and 3.1.4 show the processes of incineration and gas purification. Fig. 3.1.3 shows the incineration system schematically. Fig.3.1.2 illustrates how the combustion chamber is formed within the waste material, which consequently avoids the outer furnace walls coming into contact with molten slag. Whilst the temperature in the core is 1500° to 1600°C, the outer walls of the furnace do not exceed 60°C. Therefore no cooling for the outer walls is necessary and corrosion is widely eliminated. The waste material is evenly distributed and pushed toward the combustion zone by a revolving distributor (see Fig.3.1.2). It melts as it approaches the burner in a thin layer which is consumed between 1300° to 1400°C. The molten slag drips through the cooled bottom orifice of the furnace forming a monolithic block when collected in an uncooled crucible or as pearls when collected in a water basin. The burner can be fed with contaminated organic liquids such as tributylphosphate or used oil. To maintain the high melting temperature 30 to 60 % of excess air is injected.

The waste material is fed from a mixing silo into the incinerator by a worm-feeder. The part so far described is already operating with inactive waste.

The off-gases pass through a post-combustion chamber to ensure their complete combustion before they are directed to a heat recuperator and further to the purification system. The off-gas train is given in Fig.3.1.4. This system has yet to be built.

3.1.2.1 Method of high temperature incineration

The most common combustible and low Pu-contaminated waste, together with most of the concentration arising from processing of liquid effluent are molten in the incineration chamber of the "FLK 60" plant (at 1300° to 1400° C) to form an inert, insoluble basalt-like slag. Semi-liquid and pasty solid materials, such as chemical sludges, are pre-dried in a rotary-drum and heated by a fraction of incineration off-gases. As the properties of the slag obtained depend largely on the composition of the feed, the waste composition should be as constant as possible. To assure an utmost constant feed the solid waste is crushed into small bits and sorted in glove boxes beforehand. Together with a certain amount of dried chemical sludges it is then stored in an air-tight mixing silo from where it is conveyed through a worm-feeder into the incinerator. A heat recuperator, dust filter, chlorine scrubber and absolute filter will constitute the off-gas cleanup.

3.1.2.2 Method of moderate temperature incineration

The same type of incineration performed at lower temperatures (600° to 1200° C) will form ashes which could allow Pu-recovery. Only heavily Pu-contaminated waste will be treated in this way and no liquid waste concentrates should be added. By adding molten salts, like $Na2CO3$ and $NaSO4$, the melting point of the slag is lowered and a more soluble ash product is achieved. Such additives have the advantage of binding the HCL in situ. For this process a small scale pilot plant (see chapter 3.1.4.3) is envisaged. It will be connected to the off-gas purification system of the FLK 60 plant.

3.1.3 Range of waste

3.1.3.1 Low Pu-contaminated waste

In order to promote the slag-forming process and to achieve a highly leach resistant ash product incombustible waste materials of a minimum of 10 to 15 % mineral constituents are added to the combustible portion of the load. The contamination of the waste will be smaller than 2 g Pu per 200 l drum.

3.1.3.1.1 Combustible waste

As combustible waste are considered :

- Miscellaneous materials containing large quantities of polyethylene, polyvinylchloride, rubber, filter, swabs, rags, etc.
- Natural materials : wood, paper, cotton, etc.
- Ion exchanger : synthetic or natural,
- Sorbents : active carbon in most cases
- Organic solvents : tributylphosphate, oil, etc.

3.1.3.1.2 Incombustible waste

The incombustible portion consists mainly of chemical sludges resulting from the treatment of low-level liquid effluents by coprecipitation, application of mineral sorbents or by evaporation. Smaller quantities of crushed glass and metal items, glass wool filters, diatomaceous earth and sand from sandfilters are added.

3.1.3.1.3 Composition of waste mixture

Combustible and incombustible waste materials are collected separately. In order to obtain an optimum combustion and a convenient melting point of the slag a further subdivision of the waste material is recommended. Segregation is roughly made between :

- synthetic materials (polyethylene, PVC, ion-exchanger, etc.)
- natural materials (wood, paper, cotton, etc.)
- chemical inorganic materials
- glass and metals

A typical composition yielding a highly leach-resistant ash product is as follows :

30 % paper
10 % polyethylene
10 % ion-exchanger
10 % mineral sludges (mostly $AL(OH)_3$, $Ca_2(PO_4)_2$, $Cu_2Fe(CN)_6$, $Fe(OH)_3$)
10 % metals
10 % glass
20 % water

3.1.3.2 Heavy Pu-contaminated waste

The optimum composition rendering dissoluble ashes for maximum Pu-recovery has yet to be fund. This is intended by the experimental operation of the pilot plant (see chapter 3.1.4.4). The upper level of Pu-contamination will be determined by the maximum permissible Pu-concentration at any point of the incineration process.

3.1.4 Contents of programme

3.1.4.1 Modification of the "FLK 60" plant

The industrial plant "FLK 60" is already in inactive operation since beginning of 1975. To adapt the plant for the incineration of low Pu-contaminated waste the following modifications have to be carried out :

- Construction of an α-tight containment
- Installation of pre-treatment facilities for sorting, shredding, mixing and feeding of the waste material
- Installation of remote handling facilities
- Conversion of surroundings to a pressurized suit area
- Adaption of ash retrieval system for Pu-waste
- Installation of an off-gas control and purification system which will also cope with the off-gases coming from the later attached pilot plant (see chapter 3.1.4.3).

3.1.4.2 Operation of the "FLK 60" plant

The "FLK 60" plant will be operated to investigate the influence of waste feed composition and additives on the combustion temperature, on the off-gas production and the ash quality. Some tests will be devoted to the neutralization of acids in situ and to the removal of volatile elements in the off-gas. Various incineration conditions (oxygen supply, residence time of waste in combustion chamber etc.) will be tested. The operational studies will be supported by laboratory tests.

3.1.4.3 Construction of pilot plant

A small scale pilot plant (capacity 10 kg/h) of the same principle as the FLK 60 will be built and attached to the off-gas purification system of the FLK 60 plant. Pu-control and safeguards will be installed throughout the system.

3.1.4.4 Operation of pilot plant

The relatively small scale of the plant allows to treat higher contamination levels and gives more experimental flexibility. The main task will be to investigate the incineration of plutonium-oxychloride. Possibilities will be examined whether chlorine gases can be trapped in the ash in order to avoid secondary waste and the necessity of a titanium scrubber.

3.1.5 Characterisation of the process end products

3.1.5.1 Highly leach resistant ash product

Waste and ash samples as well as the off-gas composition will be analysed. The leaching rate and the density of the ash will be determined and solubility tests will be performed.

3.1.5.2 Easy dissoluble ash product

The ash products with a high Pu-concentration level originating from the pilot plant incinerator will mainly be tested for their solvent extraction rate of plutonium.

3.1.6 References

- IAEA-SM-207/7, "An integrated system for the conditioning of radioactive solid wastes and liquid waste concentrates" by N. Van de Voorde et. al., SCK/CEN Mol-Donk, Belgium.

FLOW SHEET HIGH TEMPERATURE WASTE INCINERATION

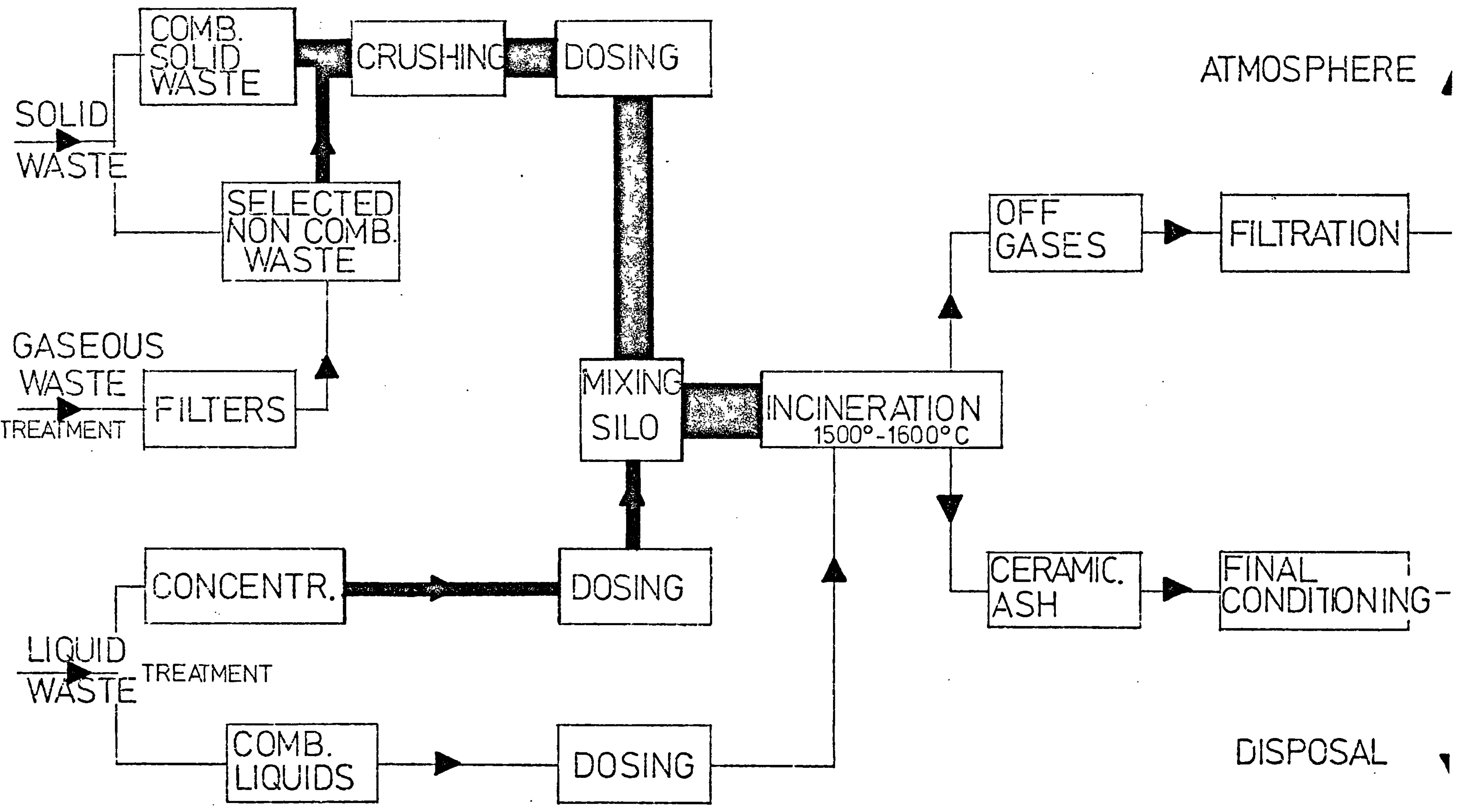
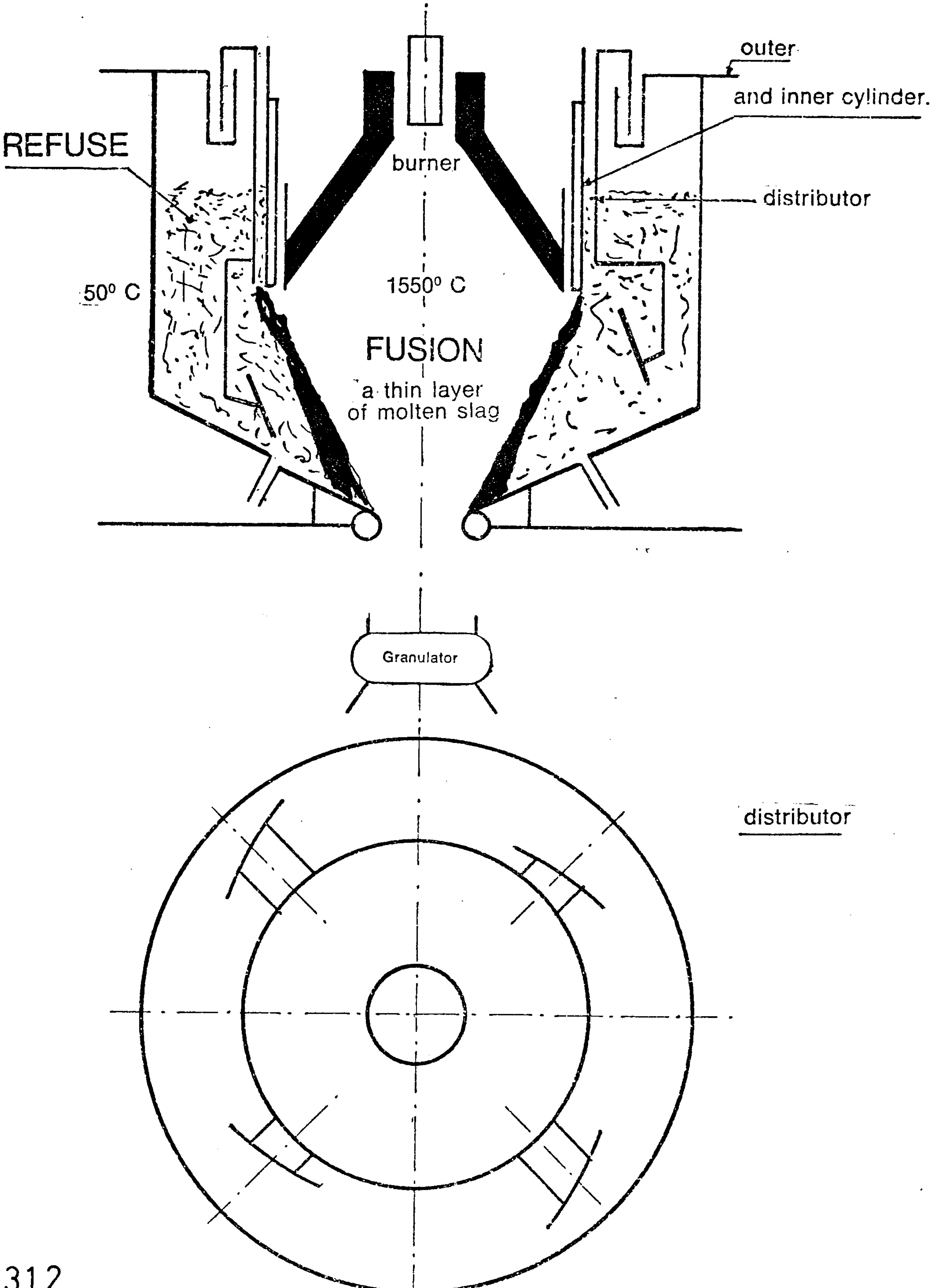


FIG. 3.1.1

DESCRIPTION OF THE FLK SYSTEM



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HIGH TEMPERATUREOVEN FOR INCINERATION OF RADIOACTIVE WASTE

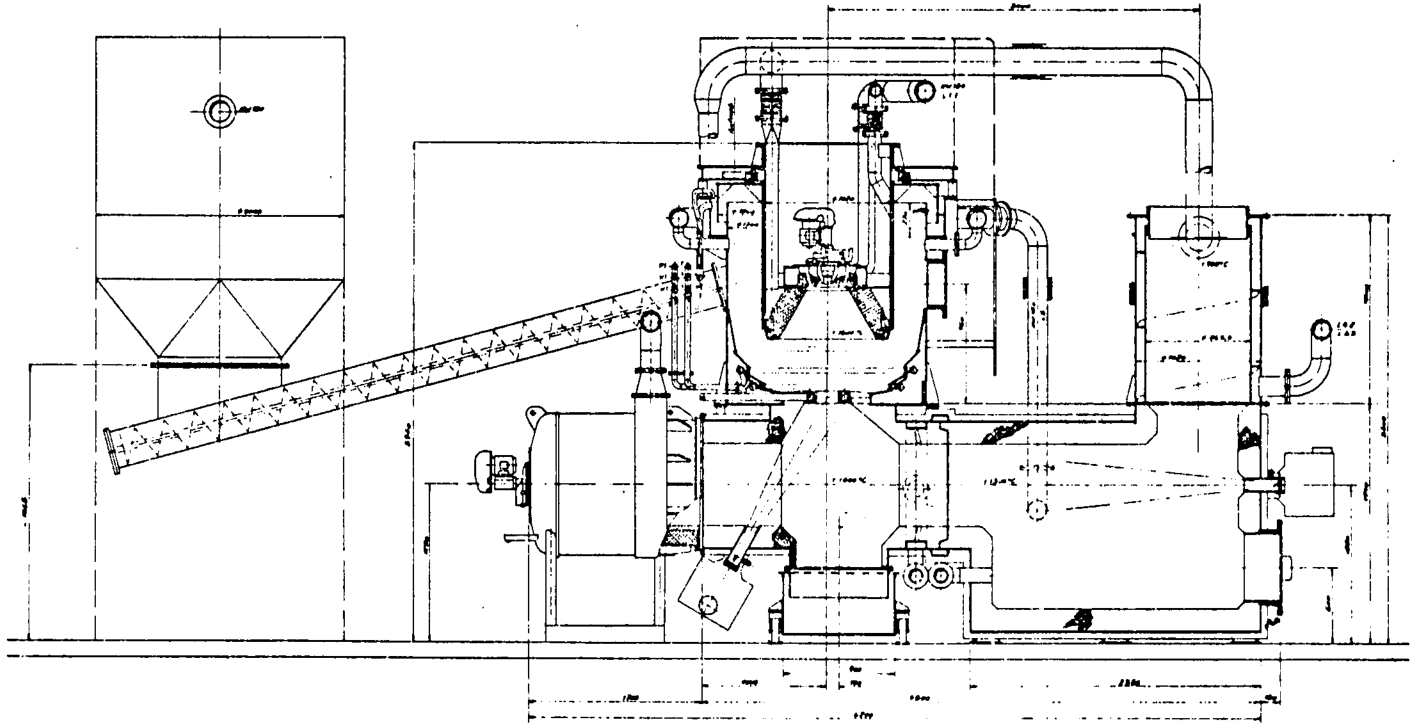


FIG.3.1.3

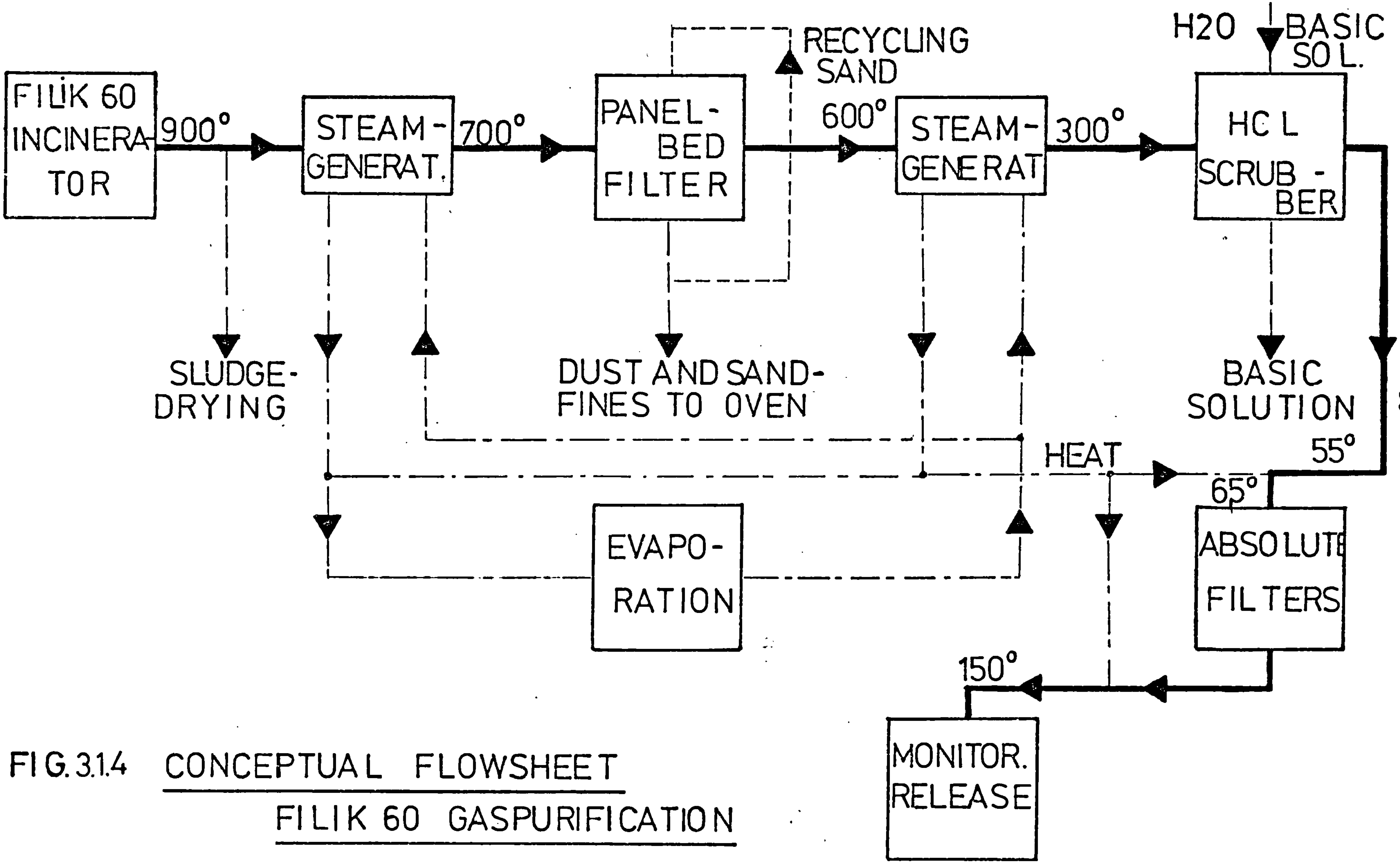


FIG. 3.1.4 CONCEPTUAL FLOWSHEET
FILIK 60 GASPURIFICATION

3.2 Acid digestion process
(NUKEM, FRG)

3.2.1 Introduction

The objective of this contract is to develop, test and optimize a pilot plant of the wet combustion method. The plant is supposed to treat 1000 kg Pu- contaminated and combustible solid waste containing approximately 10 - 12 kg of Pu at the Eurochemic plant in Mol.

3.2.2 Principle of technique

The waste is heated in a concentrated and boiling mineral acid solution. The organic compounds are decomposed and most of them are reduced to carbon. By addition of strong oxidizing agents the carbon is transformed into CO_2, which leaves the combustion chamber with the off-gas. The noncombustible portion of the waste will either be converted into the respective salts or remain inert in the acid.
The pilot plant will use H_2SO_4 as the mineral acid under addition of 5 vol. % of HNO_3 as the oxidizing agent. The operational temperature will be in the range of 230 °C to 250 °C. 70 % - 80 % of the NO_x- and SO_2- reaction gases escaping with the gaseous oxidation products are envisaged to be recycled.

Most of the plant will be built of quartz-glass components. The acid solution $H_2SO_4 + HNO_3$ will be circulated in the digester vessel.
The digestion rate will be approximately 3 kg/h. The Pu-content will be controlled at any vital point of the process (see flow-sheet Fig. 3.2.1). About 98 % of the plutonium are estimated to be recovered. The residues, converted into salts, can be treated as low active waste.

Hydrochloric, sulphuric, nitric acids and water vapours are fed to the acid recycling system. The sulphuric and nitric acids are recycled, whereas water and hydrochloric acid are treated as secondary waste.
The remaining off-gases are filtered.

The acid solution, when saturated, is drawn off from the combustion chamber, cooled in order to precipitate the salts on a revolving suction filter. The process solution will then be conducted to separation columns.

The filter cake, which contains mainly metal salts, is washed in diluted nitric acid. The plutonium will be extracted as Pu NO_3. The residues are envisaged to be incorporated into bitumen for intermediate storage.

3.2.3 Range and preparation of waste

Low Pu- contaminated waste will be treated separately (compression). A glove box serves to sort the waste into its combustible and incombustible fractions. The combustible waste to be treated consists of :

- paper, cardboard
- wood, cotton, rubber gloves
- plexiglass
- polyvinylchloride
- polyethylene
- swabs, rags, oil
- cellulose filters

No predrying of the waste is necessary. The waste will be chopped by a shredder in order to enable the blending of a fairly constant average mixture. This mixture is then fed in defined doses (plastic bags) into the combustion chamber.

3.2.4 Contents of programme

3.2.4.1 Elaboration of basic data for the combustion and Pu- recovery process

For the layout, the construction and particularly for the operation of the pilot plant, basic data have to be compiled. These studies will be mainly dealing with the digestion process, the off-gas treatment, the acid recycling and the Pu- recovery.

3.2.4.2 Engineering of the pilot plant

The pilot plant will be designed to perform the complete process as represented in the flow-sheet Fig. 3.2.1

Particular attention will be devoted at the design stage to the following :

- Pu- control within the system
- Retrieval of sulphuric and nitric acids and their vapours in the off-gas system
- Purification of the off-gas
- Recycling of the secondary waste (hydrochloric acid, water, filter, etc.) for re-combustion.

3.2.4.3 Construction and commissioning of the plant

Before assembling the plant, the following main subassemblies will be tested separately :

- Acid digestion
- Acid retrieval
- Off-gas purification

The plant will be tested in cold and hot operation.

3.2.4.4 Operation of plant

The plant will be treating ca. 1000 kg of Pu- contaminated solid waste from Eurochemic. Experiments at various operational conditions (HNO_3-supply, combustion temperature, duration of combustion process, etc.) will be carried out.

3.2.4.5 Evaluation of experimental results

The process will be evaluated according to the main parameters with respect to :

- Combustion efficiency
- Efficiency of the off-gas system
- Acid recovery rate
- Pu- recovery rate
- Characteristics of the residues.

3.2.4.6 Economy of the process

The economic aspects of a large digestion unit for the waste treatment of an industrial fuel fabrication plant will be evaluated.

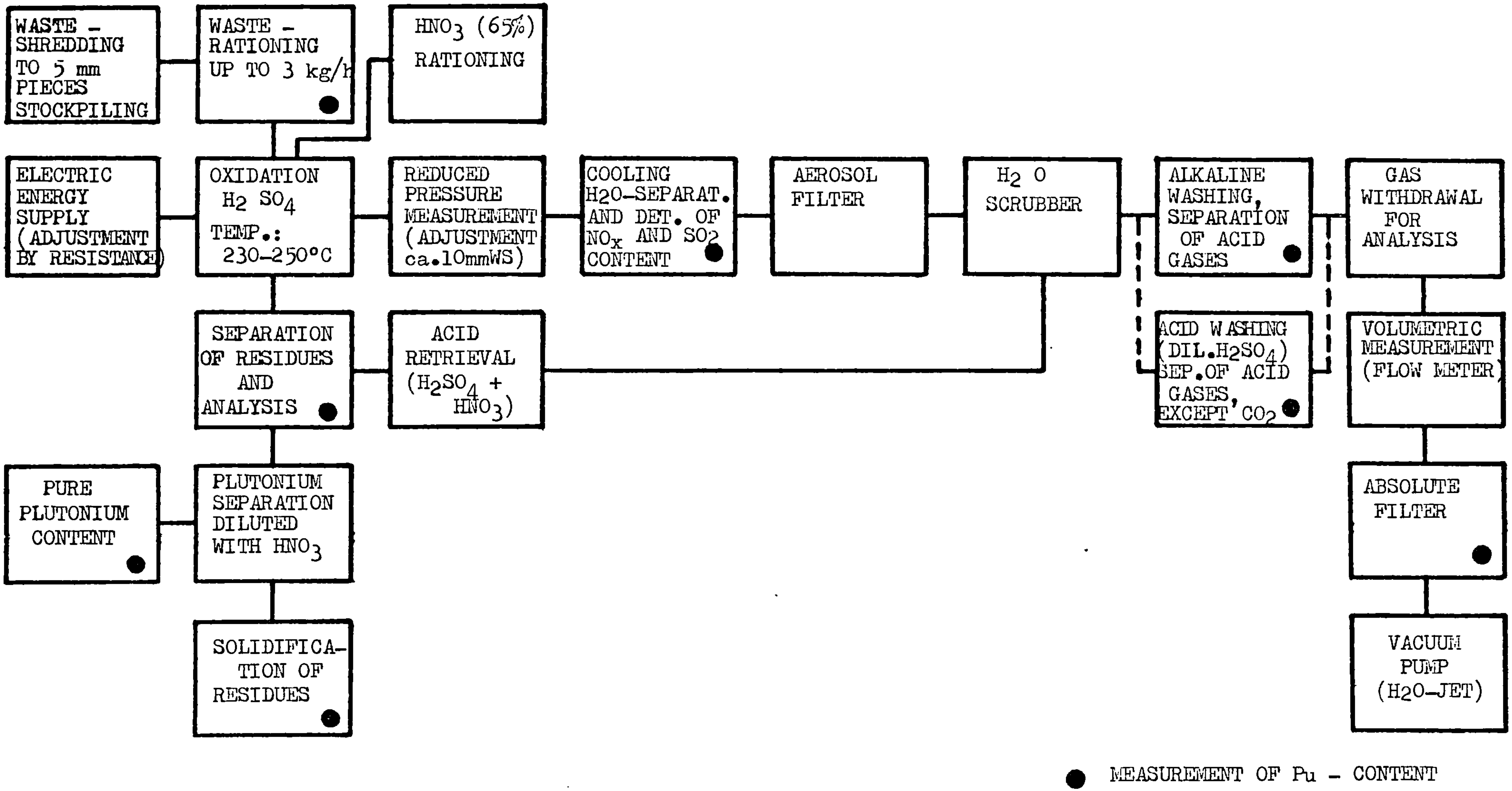


FIG. 3.2.1 : FLOW-SHEET OF ACID DIGESTION PROCESS FOR COMBUSTIBLE ALPHA-WASTE

3.3 Incineration in Molten Salts (Agip Nucleare)

3.3.1 Objective

The aim of the R & D contract with Agip Nucleare is to establish the basic data for a waste incineration technique, that permits waste incineration and Pu separation in one process step. The intention is to destroy the combustible waste in a molten salt bath with strong oxidants with the Pu remaining dissolved in the molten salts.

By filtration all the non consumed and incombustible waste would be separated from the Pu (and other heavy metals) dissolved in the fused salt. A second step would recover the Pu from the salt.
This programme is scheduled to run until the end of 1978.

3.3.2 Scope of the programme

The technique studied by Agip Nucleare, unlike Atomics International's molten salt process, will use alkaline salts 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.

This programme intends to collect sufficient data and assure adequate knowledge of the reference process to permit an evaluation of this new technique.

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

The second part of the programme consists mainly of the construction of a bench scale rig which can, on a small scale, perform the entire process and of the experimental operation of the rig with Pu contaminated waste.
The beginning of this part is subject to a favourable outcome of the first part.

3.3.3 Experiments with inactive and simultated waste

These experiments will investigate the following items :

- Choice of the most suitable salt mixture
- Determination of optimized process parameters

- Control of the concentrations of chlorides, carbonates and nitrates in the melt
- Measurement of the combustion rate and the volume reduction
- Choice of a filter for the molten salts
- Analysis and study of the treatment of the off-gas
- Measurement of the properties and analysis of the composition of the residual waste
- Conditioning of the residual waste
- Purification and recycling of exhausted or contaminated salts
- Reference data for an optimized process and
- Feasibility study of a bench scale plant.

3.3.4 Experiments with Plutonium

Using the most promising salt mixtures determined in the inactive tests, the tests with plutonium shall treat the following problems :

- Dissolving the Pu in the salt melt with acid
- Determination of the solubility and stability of Pu-compounds in the salt mixture at various temperatures
- Pu recovery techniques
- Minimization of Pu losses.

3.3.5 Part II : Construction and operation of a bench scale test rig

Based on the process characteristics resulting from the first part of the programme, a small test rig will be constructed, which will allow the following experimental investigations of the complete waste treatment process (with Pu) at a small scale (50-100 gr/h) :

- Feed and transport mechanisms, pressure balancing, avoidance of Pu deposits and combustion control
- Immersion of the waste into the molten salt, removal of incombustible residues from the reaction vessel, reduction or elimination of crud formation on vessel walls
- Determination of operating conditions with least incombustible residues and least risk to the off-gas filters
- Influence of the combustion products upon the Pu extraction process.

For the experimental operation with Pu the rig will have to be fully contained and operated on an appropriately licensed site.

4. PROPERTIES OF SUBSTANCES SUITABLE FOR IMMOBILIZING HIGH LEVEL WASTE

JOINT PROGRAMME UKAEA - HMI - CEA

Testing and evaluation of the properties of various potential materials for immobilizing high-activity waste in a solid form.

4.1 Introduction

This programme is a coordinated action of the United Kingdom Atomic Energy Authority "UKAEA" (Great Britain), the Hahn Meitner Institut "HMI" (Germany) and the Commissariat à l'Energie Atomique "CEA" (France). The objective is to establish a joint basis for the evaluation of those solidification substances which will be applied in the near future. The programme will be centred on series of experiments jointly testing five different solidification substances from the three participating countries, performed under strictly identical conditions. The experimental investigations will include the following main items :

- Determination of the leaching rate ;
- Investigation of the temperature effects ;
- Investigation of the alpha-radiation effects.

The three coordinated contracts are scheduled as follows :

UKAEA : Between 1st January 1977 and 31st December 1979.

HMI : Between 1st October 1976 and 31st December 1979.

CEA : This contract, which will have a maximum duration of six months, will be negociated in the first half of 1977.

4.2 Scope of programme

4.2.1 Determination of leaching rate

4.2.1.1 Block leach test on active samples

CEA will measure the leach rate of five active block samples with volumes of approximately 1 litre each containing up to 20 % of light water reactor fission product oxydes (activity 100 to 1000 Ci/$\beta\gamma$).

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 : Cs 137, Sr 90, Ce 144 and Ru 106,
- 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.

4.2.1.2 Leach test on small inactive samples

A corresponding series of Soxhlet type leach rate measurements will be carried out by the UKAEA on small inactive samples simulating the reference waste products.
These leach tests at standard conditions provide a base line for the leach tests measuring temperature and radiation effects. Five specimens of each reference substance will be tested. The main characteristics of this measurement technique are :

- Specimen form :	coupon, 1 x 10 x 10 mm
- Leaching agent :	pure water
- Temperature :	100°C
- Duration of test :	2 to 20 days depending on leaching resistance of sample
- Water/solid ratio :	10/litre to 30/litre
- Water exchange rate :	about every 10 minutes.

The leach rate is measured by the total weight loss. A second series of tests will be carried out using the base line conditions (as above) but under reduced pressure and at differing temperatures. One specimen of each substance will be measured at :
50°C, 60°C, 70°C, 80°C and 90°C.

4.2.1.3 Supplementary leach tests on inactive samples

These include two series of measurements which will be carried out by HMI on inactive samples containing simulated waste.

4.2.1.3.1 Granulate titration method

This method measures the alkali loss according to DIN 12111.
The conditions of this test are :

- Specimen form : powder (particles of 2g)
- Leaching agent : pure water
- Temperature : 100°C
- Duration of test : 60 minutes
- Water exchange rate : 100 ml/h.

4.2.1.3.2 Phase selective leaching

The objective of this test is to determine the leach resistance of crystal phases and of the remaining vitreous phase on polished surfaces of devitrified glasses under the following conditions :

- Specimen form : coupons (ca. 100 mg)
- Leaching agent : optional
- Temperature : 20°C - 100°C
- Duration of test : optional.

4.2.2 Investigation of temperature effects

This study investigates the changes likely to occur in the physical state of waste glasses intended to confine radioactive waste for up to 1000 years. These very slow changes from a fully vitreous to a partially crystalline structure will depend on the composition of the glass and the storage temperature. Negative effects upon the retention of the waste glasses are only anticipated in the long term and the effects of partial devitrification are not necessarily negative.

However, as the vitrification of radioactive waste is almost irreversible, the long term characteristics of the waste glasses must be investigated before relying on them to retain the waste for the next centuries. In order to accelerate the devitrification, the glasses are annealed at higher temperatures (500°C to 800°C) and exposed for varied annealing times (hours to months). The long term properties will be determined subsequently.

4.2.2.1 Heat treatment of samples

Glasses held at elevated temperature while cooling from the formation temperature propagate devitrification effects. Two sets of identically pretreated samples, one for UKAEA, one for HMI, will be prepared by UKAEA. Each set consists of one untreated control sample and for each of the following annealing temperatures one sample per annealing time given below.

Annealing temperatures : 500°C, 600°C, 700°C and 800°C

Annealing time : 2 hours, 1 day, 10 days and 100 days.

Leach tests will be performed according to chapters 4.2.1.2 and 4.2.1.3 on samples of each variant. The remaining samples will undergo the following investigations.

4.2.2.2 Investigation of structural integrity

These investigations performed by HMI complement the UKAEA efforts (s.Chap.4.2.3) in the field of devitrification by analysing the extent and the composition of the crystalline phases and by measuring the retention properties of these phases in respect to the parent glasses by phase selective leaching. This could give very valuable information on how to improve the glasses, it could explain abnormal results and could supply further evidence and general knowledge about devitrification phenomena. The programme provides the following investigations :

- Investigations of softening behaviour, form stability and devitrification ;
- Time and temperature dependent behaviour of (simulated) fission products in the substances after thermal treatment ;
- Detailed investigation of the structural changes produced by thermal treatment (degree of crystallization, type and size of crystals, crystal growth rates).

4.2.3 Investigation of alpha-radiation effects

The effects of alpha-radiation are considered to cause the most significant radiation damage in waste glasses. The UKAEA will examine the effects of alpha-radiation upon four specimens doped with 238 $Pu\ O_2$ of each of the test substances after having stored these substances for two years. During this time they will receive an irradiation dose of about 2.2 x 10^{18} alpha-decays per ml. A set of simular specimens containing 239 Pu O_2 will be made as controls. A schedule of the tests to be performed on the respective specimens is set out in Table 1.

Table 1 : Alpha damage tests

Specimen Number	Holding Temperature	Test after holding
1	Room temp.	(Wigner) Energy storage
2	Room temp.	Leach rate, crack testing, microstructure
3	150 °C	Helium release, leach rate
4	300 °C decreas-sing to 40 °C	Leach rate, crack testing, microstructure

4.3 Equipment for measuring and testing the required properties

The necessary equipment to carry out the envisaged programme is largely available at the three laboratories. Table 2 gives a survey of the test methods which will be employed.

Table 2 : Test methods

Test	Laboratory	Method
Block leach test	CEA Marcoule	leaching, radioanalysis
Soxhlet leach test	UKAEA Harwell	leaching, weighing
Supplementary leach tests	HMI Berlin	leaching, pH-measurement, microscopy
Structural integrity tests	HMI Berlin	differential thermal analysis, differential thermal gravimetry, screening electron microscopy, x-ray diffraction, electron beam microanalysis, optical microscopy
Alpha-damage tests	UKAEA Harwell	differential thermal analysis, microscopy, mass spectrometry, ultrasonic crack detection

4.4 Reference substances

The five reference substances to be investigated are given in Table 3. The Celsiane glass ceramic B 1/3 was developed at HMI and the borosilicate glass G 98 at GfK - Karlsruhe (D1 and D2 in Table 3). The other four substances are also borosilicate glasses. The second glass of the two French glasses (F1 and F2) is suitable for HLW containing ca. 6 % of Gd_2O_3 and a similar fraction of iron oxides. All the samples, apart from those reserved for the block leach test, are manufactured and pretreated (annealed and doped) by the UKAEA. The block samples are manufactured in the Vulcain facilities in Marcoule (CEA).

Table 3 : Reference Substances

Tested by	Substances					
	D1 Glass Ceramic	D2 G98	GB1 209	GB2 189	F1 SON 58 30.20.u2	F2 SON 64 19.17
HMI	x	x	x	x	x	
UKAEA	x	x	x	x	x	
CEA	x	x	x		x	x

The French glass F2 will only be tested by CEA replacing the English glass GB2.

4.5 Coordination of the programme

In order to coordinate the programme and to exchange experience between the three participants, joint progress meetings will be held at about six months interval. Representatives from other organizations within the Community, who are engaged in similar or related activities will be able to participate at these meetings. One of the tasks of this working party will be to discuss and recommend for further consideration possible research activities to be incorporated in future EC programmes.

References

- IAEA-SM-207/24 "Development and radiation stability of glasses for highly radioactive wastes" by A.R. Hall et. al., AERE Harwell Didcot Oxon G.B.
- IAEA-SM-207/18 "Evaluation of products for solidification of high level radioactive waste from commercial reprocessing in the Federal Republic of Germany" by E. Ewest and H.W. Levi, Hahn-Meitner-Institut für Kernforschung Berlin GmbH, Fed. Rep. of Germany.

- IAEA-SM-207/36 "Confinement de la radioactivité dans les verres" by F.R. Laude et. al., Comissariat à l'Energie Atomique, Centre de Marcoule, F.

5. ENGINEERED STORAGE FOR SOLIDIFIED HIGH LEVEL WASTE

5.1 Scope of the programme

Although a wide variety of storage designs can be imagined the programme should be limited to three basic concepts : the water cooled pond concept, the air cooled concept and a mixed system, which would provide a water cooled pond during a first period of decay heat removal and air cooling during the remainder of the total storage time.
In order to obtain comparable data on cost and safety of the storage systems, the study should be based on a set of common reference data, construction codes, provisional design safety criteria and a common costing system, which would be agreed upon in meetings of the participating firms : Belgonucléaire, Nukem, Agip Nucleare.

5.2 Stage I : Common Reference Data and Options

The appended logic diagram presents a scheme for the integration and coordination of the programme.
Although the sequence of actions is, at least during the first two stages, determined by the requirement to establish a number of alternative conceptual designs as reference designs, the following sequence of economic and safety study of the concepts can readily be inverted or arranged in parallel.
The first stage begins with the selection of a coherent and realistic set of input data and limiting options. The list at the head of the diagram gives a number of main options and data, upon which the study should be based :

1) General terms of reference :
   - In the waste management strategy, the storage should be followed by geological disposal
   - Duration of the storage : 50 years ?
   - Annual capacity of the reprocessing plant
   - Hypothetic site conditions

2) Spent fuel reference data :
   - Type of fuel : (UO_2, PWR)
   - Enrichment
   - Burnup
   - Precooling time

3) H.L. Liquid waste reference data :
   - Reprocessing technique (Purex)
   - FP and actinide inventory of liquid waste
   - Heat generation VS. time
   - Chemical composition (reference data)

4) Tank storage reference data :
   - Risks of tank storage } for typical tank form
   - Costs of tank storage }

Starting with the acceptance of this set of common reference data and other terms of reference, two strategies should be examined

a) Vitrification immediately after reprocessing
b) Vitrification after n years of tank storage

The design concepts for storing the solidified HLW under the two strategies would be

1) The water cooled (pond) concept
2) The mixed (first water - then air cooled) concept
3) The air cooled concept

For strategy a) (no tank storage), transferring the solidified waste from briefly stored LWR fuel with 30000 MWd/t directly to an air cooled storage does not appear very promising. This possible alternative has therefore been omitted from the scheme.
The target of the first stage is to establish the basic elements of design for the three concepts. These elements would comprise : design data for heat removal systems, storage volume, layout of storage and handling systems, draft schemes for electrical and other supply systems and other basic design information.

5.3 Stage II : Design Study of the Storage Systems

The second stage of the programme should begin the establishment of the safety criteria and the design and construction codes.
These common criteria and design rules would only be accepted for application within the design and evaluation study of this programme and an agreement within the framework of this study would not be binding at a later stage of the Commission efforts to establish recommendations or common criteria on this matter.

The assumed common criteria would, among other items define :

- to what extent the design should take into consideration earth quakes, aircraft crashes, sabotage or floods ;
- what maximum temperatures in the vitrified waste and the structures should be admitted for routine and non routine conditions ;
- which degree of redundancy should be provided for vital equipment and engineered safeguards ;
- characteristics of the hypothetical sites relevant to safety assessments.

The next step in the programme will be the study and design of the waste containers. Once the waste containers have been defined, the conceptual design of the storage plants could proceed. At the end of Stage II, the conceptual designs are frozen.
The conceptual design studies should now contain all the elements necessary to assess the risks of the five versions and to estimate the cost of building and operating them.
The amount of detail required for the design work should be determined by the requirements

a) of the fault-tree type risk analysis,
b) to produce a reasonably complete breakdown of major plant components and subassemblies for the investment cost estimates and
c) to supply the necessary data for assessing the operating cost, i.e. consumption of electricity and water, the number of personnel, the expected maintenance frequency, component lifetimes and other relevant items.

As mentioned previously, the first step towards assessing the risks of the various designs, the establishing of the sequences of events of fault trees which could lead to a danger situation, could begin at this stage. This has not been presented in the diagram to avoid confusion.

5.4 Stage III : Evaluation and Analysis of the Results

Having established the conceptual designs, a third progress meeting will establish a system which will allow a straightforward comparison of the economical aspects of all versions.

This system could be based on a common method of costing and compiling costs as well as a common set of prices for building and construction materials, major plant components, manpower, etc. as well as common financing conditions.

The contracting firms should, however, be able to produce cost estimates for building and operating the plant, which will not reflect differences in national prices and cost structures, but will with reasonable accuracy permit comparison between the five versions.

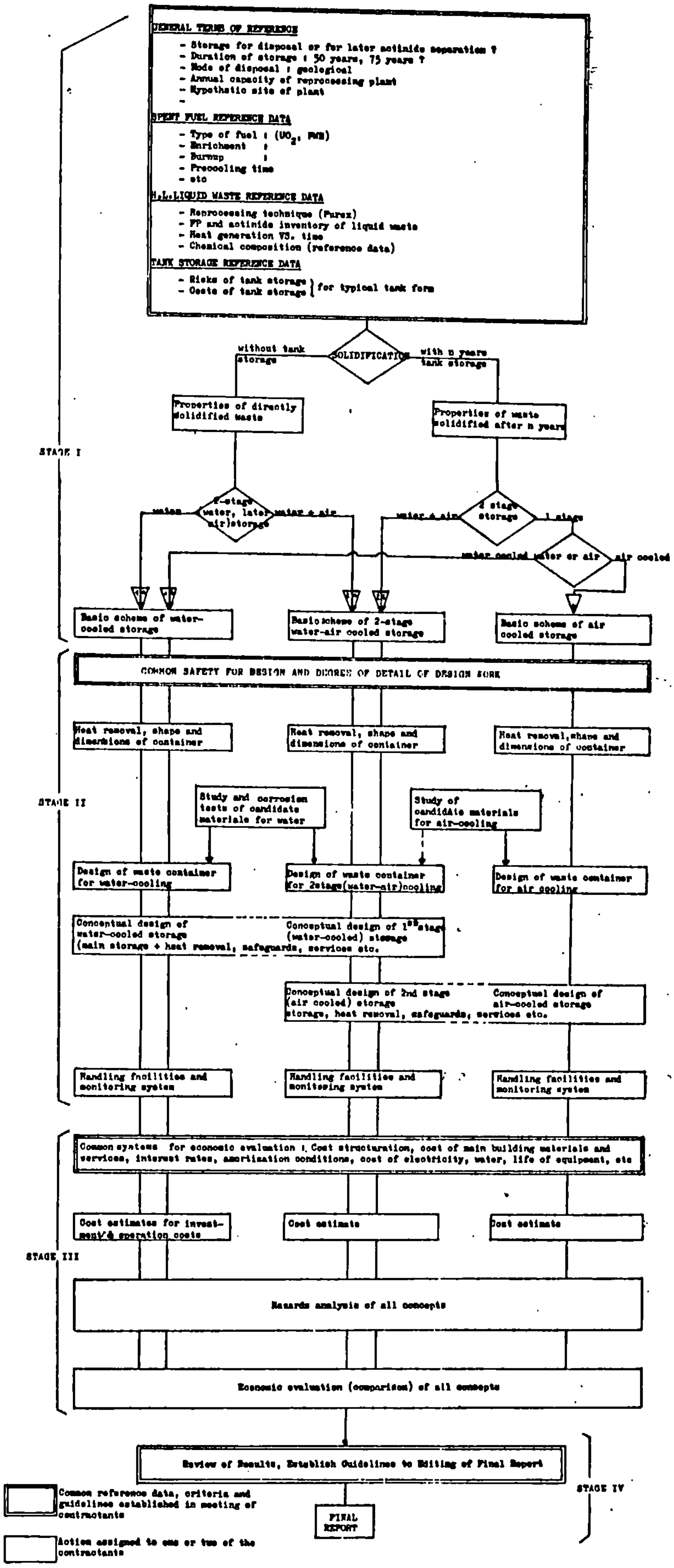
The design studies will then be submitted to a risk analysis.
For this purpose fault trees of all initiating events which could lead to releases of radioactivity will be established and the probability of a release will be assessed.
The result will be evaluated in a former type analysis.

The probabilistic analysis will show up the weakest spots in the engineered safeguards, permit a comparison of the variants and quantify the risk of storing solidified HLW.

Stage IV of the programme consists of a review and the elaboration of the joint final report.

STUDY OF ENGINEERED STORAGE

Programme Coordination



6. LONG TERM STORAGE AND DISPOSAL OF SOLIDIFIED HIGH LEVEL AND/OR ALPHA-ACTIVE WASTE IN GEOLOGICAL FORMATIONS

6.1 Catalogue of geological formations suitable for the disposal of radioactive waste

The catalogue will provide an overall view of the geological formations of potential interest for the disposal of high level and/or -bearing waste in the Member states.

Participating institutes :

- Service géologique de Belgique
- Danmarks Geologiske Undersøgelse
- Bureau de Recherches géologiques et minières (France)
- Direzione generale delle Miniere, Servizio geologico (Italy)
- Rijksgeologische Dienst (Netherlands)
- Bundesanstalt für Geowissenschaften und Rohstoffe (Federal Republic of Germany)
- Institute of Geological Sciences (United Kingdom)
- Geological Survey of Ireland.

Denmark's and Netherlands' contributions will be free.

A joint scheme has been agreed upon to insure.

1) that the national studies will be carried out on a comparable basis using common criteria

2) that the results, given on an European map (scale 1/1 500000) will show the same degree of information and use the same symbols.

First draft contributions to the catalogue are expected for the end of 1977. The final version of a complete harmonized document will be published at the end of the programme.

6.2 Storage in Salt Domes

6.2.1 Objectives

This programme conducted by GfK (Gesellschaft für Kernforschung mbH) aims at the construction of an experimental site and its handling equipment as well as the development of a transport container for a storage experiment in the Asse salt mine. This experiment will use genuine highly active glass blocks as produced by the solidification of High Level Waste.
The storage experiment will effectively start in 1984 at the latest - earlier, if the required licensing permits.

6.2.2 Programme

6.2.2.1 Laboratory and in-situ experiments

These experiments will provide the necessary knowledge for the assessment of the conditions (temperatures, mechanical loads, humidity, radiation) in a storage vault and their influence on the stability of the site configuration and the properties of construction materials.

Two distinct types of experiments are envisaged :
The differential measurements of certain material parameters e.g. the thermal conductivity of the salt in a laboratory and the integral in-situ measurements of the heat removal in the Asse mine, which will strive to simulate the actual storage conditions as closely as possible. For various reasons only an appropriate simulation can be achieved. Flexible computer codes are therefore required to forecast in particular the temperature transient and the resulting geomechanical consequences. The input of this code will consist partly of the state dependant material parameters determined in the differential measurements. On the other hand the integral in -situ tests are indispensable for the verification of the computer codes.

6.2.2.2 Measurement of the temperature distribution and convergence measurements

These measurements will be made in lined and unlined boreholes using electic heaters. The following actions are planned :

- preparation of the experiment area and assembly of the experimental equipment,
- development of a standard probe for systematic investigations of heat - induced borehole convergence,
- convergence measurements in drifts and boreholes in the context of thermal tests,
- establishment of X - ray laboratory methods and the elaboration of a study for the determination of shear stresses in the rock.

6.2.2.3 Complementary Tests

The following experimental investigations are envisaged :

- determination of the rheological properties of the Asse salts at elevated temperatures and after irradiation
- corrosion study of materials used for borehole lining and waste conditioning in salt at higher temperatures
- tests of the migration and release of liquid inclusions in the older salt formations.

6.2.3 Computer Codes

A number of computer codes will be required for the final design of the scheduled experimental storage and for a large repository of radioactive waste envisaged at a later stage.
With the aid of these codes it will be possible to predict events with relevance to the safety notwithstanding the complex structure of the storage site and the properties of the salt which can only be determined and described with difficulty.
The following actions are planned :

- Improvement of the available codes for calculation of temperature distribution in glass and salt.
  These codes cover the conditions for the different arrangements of boreholes as well as these in a wider space in the salt dome and the rock above it.

- Development of codes for calculating the stability of individual pillars and of the entire storage vault, taking the temperatures into consideration.

- Development of calculating methods to predict the consequences of a water or brine irruption into the mine in view of the heat released from the waste (convection, mass transfer).

6.2.4 Technical installations and equipment

The technical development effort for the transport, loading and monitoring equipment will concentrate on :

- a shielded flask which will satisfy all licensing requirements for transport and handling of full size active waste blocks
- development and acquisition of other technical equipment for the transport, loading, retrieval as well as monitoring of the stored waste glass.

6.3 Storage in crystalline formations I

6.3.1 Objective

The aim of this project of the CEA (Commissariat à l'Energie Atomique), which will be carried out in 1976 and 1977, is to gather all the data required for the evaluation of the ways and means of storage in a crystalline environment. Synthesis of this data will enable a selection to be made of rock formations suitable for receiving the waste. On one of these formations, prospecting methods will be developed for the purpose of :

- determining areas with the most favourable characteristics for the confinement of radioactive waste storage ;
- developing a series of models to simulate the main transfer phenomena liable to bring out a return of radioactivity to the surface environment from a crystalline rock site ;
- studying the geochemical phenomena (adsorption) that may occur after leaching by underground water when radioactive waste is stored in a crystalline environment.

6.3.2 Selection and survey of a crystalline rock bed

This study comprises :

- the setting up of a file on the basis of a bibliographical survey of the existing data on crystalline rocks ;
- large-area geological prospecting in one of the rock formations selected, and taking from one to three favourable zones. Methods of exploration will be developed as follows :
  - a study of the surface geology aimed at defining the nature of the rock formation, its structure (mainly with respect to depth) and the degree of deformation (mapping of the petrographic facies and structural elements, followed by a photogeological study) ;
  - a geophysical study to determine the shape of the interface between the rock bed and the enveloping strata (two seismograms, reflection and refraction to a depth of 1,500 m) and the type of granite in the rocks (gravimetric and magnetic studies) ;

- investigation by sounding to check the homogeneity and in-depth structure of the granite (bore 1,000 m in length).

  The following studies and measurements will be carried out during the boring process :

  - hydrogeological, thermal and geotechnical parameters ;
  - petrographical and geochemical studies of the materials sampled ;
  - characterization and petrophysical study of the materials sampled ;
  - diagraphics ;
  - selection of the most favourable zones inside a rock bed.

6.3.3 Study of the mode of transfer of radioactive products in the geological environment : modelling of migration in the soil

This requires the development of a theoretical model representing transfer in a porous medium and taking account of the phenomena of pure convection, kinematic dispersal and molecular diffusion, as well as adsorption-desorption and the radioactive decay of the element under consideration.

6.3.4 Verification of the analytical model

The model will be truth-tested by means of :

- a test in situ with non-adsorbable tracers (verification of its representativeness with respect to convection and dispersion) ;
- experiments on cores sampled on the previous site ;
- a test in situ with adsorbable tracers (checking of the conditions for extrapolating the representation of the adsorption phenomenon to the scale of the ground).

6.3.5 Study of the artificial and natural geochemical barriers in a crystalline environment

The following geochemical barriers will be studied

- the first barrier (artificial) is the one built around the storage site of the glass immobilizing blocks and consisting of minerals suitable for immobilizing the water-soluble species of the radioactive elements ;

- the second barrier (natural) consists of the minerals contained in the crystalline rocks, in particular the hysterogenic minerals in the cracks through which the water preferentially circulates ;
- the third barrier (natural) is formed of rocks produced by wheathering of the crystalline formations, such as the granite or gneiss sands- fossil or recent - which often surround these crystalline rocks in beds of considerable depth.

The objective of the study is to select suitable materials for the artificial geochemical barrier and to forecast - by experimental simulation and computer modelling - the long-term geochemical behaviour of the long-lived radioactive elements during their passage through the three barriers described above.

The chemical elements examined will be radionuclides with a long enough half-life which are present in the waste and for which the water-soluble species found in natural solutions will be determined.

Experimental research will be carried out to determine :

- the physico-chemical characteristics of the materials and solutions ;
- the interactions between solutions and materials during statistical or capillary-column experiments to determine the chemical changes in the major elements of the solution and the adsorption of the radioactive elements.

The outcome of the study will be a computerized simulation comprising :

- the simulation of adsorption phenomena ;
- validity tests on various mathematical models ;
- the determination of the numerical values of the various parameters on the basis of the experimental results.

6.4 Storage in crystalline formations II

6.4.1.0 Objectives

The second action investigating storage in hard rock is undertaken by the UKAEA. Its principal aims are :

- Identification of sites in hard crystalline rock within Great Britain at which the disposal in geological formations appears feasible on the basis of desk studies and field investigation. The programme will concentrate on geological and associated surveys.
- Studies of the physico-chemical compatibility between the wastes, their containers and the host rocks
- Engineering requirements or procedures for disposal and development of monitoring techniques as may be needed in the site selection surveys and demonstration experiment.

6.4.2 Programme

6.4.2.1 Geological survey and assessment of sites

This assessment will involve data collection at a chosen site and initiation of long-term monitoring procedures.

The following geological investigations are envisaged :

a. Geological survey
   - Drilling of 4 to 6 boreholes to a depth of about 100 m
   - preparation of a geological map of the site and the area within a radius of about 5 km, included a detailed joint and structural survey in the vicinity of the site

b. Petrological and geochemical survey in order to define the chemical and mineralogical variations to be expected in the host rocks and predict conditions during their formation :
   - Detailed petrographical, mineralogical and geochemical analyses of cores and field samples
   - determination of ion-exchange capacities
   - laboratory and field determination of the distribution coefficient of movement of critical nuclides

c. Hydrological survey in order to define the intergranular fissure and mixed flow components in both superficial deposits and bedrock and to evaluate any possible risk of movement of wastes by circulating fluids

d. Seismic survey (desk and field studies)

e. Geophysical survey

f. Geotechnical survey in order to solve design problems in the host rock selected (determination of basic rock properties such as thermal and hydraulic conductivity, creep characteristics, deformation characteristics, etc.)

g. Initiation of the monitoring programme
At an early stage in the investigation it is essential that a start should be made in the long-term monitoring of relevant parameters. Monitoring will initially include :

- water levels in selected wells and boreholes
- spring and stream discharge rates
- hydrochemistry, including isotope chemistry
- seismic activity
- in-situ strain and creep measurements.

6.4.2.2 Engineering studies

6.4.2.2.1 Heat transfer and physico-chemical studies

Fission product heating in the disposal waste will raise the temperature of the surrounding rock ; the study will determine both the temperatures which will be produced by varying conditions and the maximum values which are safe. The investigation foresees :

- Computation of analytical solutions for transient conduction from finite heat sources into an infinite medium
- measure of the effective physical properties from large samples
- simulation of rock disposal with electrical heating within a field experiment.

6.4.2.2.2 Engineering conceptual studies

- Optimisation of engineering schemes .
  Design studies will be made to assess the practicability and approximate costs of various engineering schemes for disposal including a single deep large cavity and an array of holes into each of which single or small numbers of blocks are dispersed throughout a large volume of rock ;
- Lining of cavity walls :
  The merits of possible lining will be assessed by heat transfer calculation and adsorption experiments.

6.4.2.2.3 Water dating techniques

- If groundwater is found in the vicinity of a possible disposal position, its age and the possibility that it could flow back to man's environment after contamination with radioactivity must be investigated. Such dating and deduction about its flow can in principle be made from isotopic measurement of the $^{234}U/^{238}U$ and $^{230}Th/^{234}U$ ratios. To extend these measurements to ranges of interest of $5 \cdot 10^4$ and $1 \cdot 5 \cdot 10^6$ years respectively will require some further development of the established technique.

6.5 Storage in clay formations

6.5.1 Objective

The maximum aim will be to place in service an experimental underground cavity in a clay formation at the CEN site at Mol, where an extensive range of in situ trials will be carried out, together with pilot-scale operation of the radioactive disposal site. The programme up to 1977 includes :

- local geological prospecting and the relevant soil analyses ;
- geotechnical and hydrological prospecting and the relevant geological characterizations ;
- reliability studies.

6.5.2 Analyses of Boom clay samples

In 1975, an initial geological drilling to a depth of 580 m produced a stratisgraphic cross-section which revealed that the argillaceous stratum at Boom (at a depth of 160-270 m) was homogeneous and possessed good characteristics for the possible establishment of a storage site. The analyses are to be performed on samples (one every five meters in the Boom clay) of the geological drilling mentioned above.

Other investigations to be carried out are :

- quantitative chemical analyses ;
- mineralogical and petrological analyses (determination of non-argillaceous minerals and the proportion of argillaceous minerals present) ;
- the determination of certain physical and mechanical properties ;
- the determination of ion-exchange properties : the coefficients of distribution are to be determined in respect of the radionuclides SR^{85}, Cs^{134}, Eu^{154}, Pu^{239}, Pu^{240} and Am^{241} on clay samples heated to various temperatures and/or previously subjected to different doses of gamma rays.

Representative tests will then be performed, taking account of the mixtures of elements present in different concentrations in the fission product solutions, the rate of leaching by underground water of the fission products and conditioned actinides and the temperatures and radiation doses to which the argillaceous masses might be exposed.

Supplementary measurements are to be made in the laboratory, namely : measurement of the natural radioactivity and qualitative determination of the radionuclides present in the various geological formations encountered ; determination of any gaseous products that might form when the clay is heated in the presence of atmospheric oxygen ; determination of the radiolysis products resulting from the irradiation of the clay by an intense gamma field and determination of the geological age of the argillaceous formations by measuring the natural radionuclides.

6.5.3 Geotechnical drilling, hydrological prospecting and the relevant geological characterization

6.5.3.1 Geotechnical drilling is designed to provide additional information on the mechanical properties of undisturbed clay samples, i.e., such as have not suffered deformation during sampling ; for this purpose, special coring equipment has been developed. The drilling has been made to the base of the Boom clay, i.e., to a depth of approximately 260 m.
Samples taken from the clay at intervals of two meters will be examined to determine their density, permeability coefficient and shearing strength.

6.5.3.2 Two additional shafts have been sunk for the installation of tubes to measure the water level and water pressure ; in this way information will be gathered on the characteristics of water-bearing beds. Five such measuring tubes will be installed at the following depths : 580 m in the geological bore, 450, 325 and 275 m in each of the two additional shafts, and 170 m in a shaft already existing.

The measurements will be made at regular intervals of time in order to chart the variations dependent on atmospheric conditions (temperature, barometric pressure, precipitation and snowfall). The measurements at the site will be supplemented by any information that may be collected from other shafts in the area, and by a careful study of the general hydrogeology of the area.

6.5.4 Location of faults

The faults will be located by geophysical surface techniques and/or by additional drillings and underground geophysical measurements. For every injection a dozen drillings could be made to a maximum depth of 100 m ; at the end of each drilling, cores of earth of a few metres in length will be sampled and examined in the laboratory, and geophysical measurements will then be made by sounding in each borehole.

6.5.5 Development of mathematical models

These models are intended to evaluate the importance of the phenomenon of ion migration underground and the influence of the thermal effects of high-activity waste on the storage concept.

6.5.6 Reliability studies from the point of view of environmental hazard and technical installations

6.5.6.1 Preparation of a report on the ways and means of disposing of radioactive waste in an argillaceous formation in Belgium

At this stage of the work the task will be to :

- collate and interpret the experimental results obtained, and fit them into the general pattern of existing information gathered together for the purpose ;
- draw preliminary conclusions with respect to environmental safety and the possibility of setting up an industrially operable plant.

6.5.6.2 Preparation of the technical specification

The following studies will be required for the purpose of drawing up the specification :

a) a general design study outlining an industrial plant for the disposal of the various types of waste under consideration ;

b) a detailed study and cost estimate regarding the construction of an experimental cavity with a volume ranging from 100 to 1000 m^3.

7. STORAGE OF GASEOUS WASTES

7.0 General introduction

Gaseous and volatile radionuclides are generated in nuclear reactors as binary or ternary fission products and as activation products. Krypton-85, tritium, iodine-129 and carbon-14 are those which may constitute important long-term sources of irradiation to the biosphere.
The mentioned radionuclides may be released at the reactors, at processing plant of spent fuel and at waste treatment facilities.
Discharge of these radioactive effluents to the environment is limited by recommendations and by regulations. There is a tendency towards more restrictive decontamination for the effluents, particularly, in the new installations. Required decontamination factors (DF), based on the recommended maximum doses for releases (MD) and planned nuclear installations, are given in U.S.A. by the Environmental Protection Agency (EPA) and in Fed. Rep. of Germany by Radiation Protection Commission (GRPC). These values are reported in the table below.

Table 1 : Max. Doses for Release MD and required Decontamination Factors DF

Organism	Iodine-129		Krypton-85	
	MD	DF	MD	DF
EPA	5 mCi/GWe•y	180	50,000 Ci/GWe•y	6
GRPC	2,4 mCi/GWe•y	340	20,000 Ci/GWe•y or 1 MCi/yr/1500MT plant	15

For tritium a DF = 7 was proposed to the GRPC.

7.1 Outline of the programme

The planned off-gas management schemes in the Member states were examined to identify needs to improve or develop alternative methods.

7.1.1 Krypton storage in pressurized cylinders

For the long term storage further investigations concerning :

- effects of the decay product rubidium and of impurities on the long term integrity of the container
- temperature distribution within the container
- design of the storage facility, considering casual cylinder failure

are needed.

The Krypton may be present in the cylinder as free gas or incorporated in a sorbant such as activated carbon.

7.1.2 Sea disposal of Krypton

A paper study on this subject shall be done covering the following aspects :

- feasibility and design ;
- safety analysis of the different steps ;
- behaviour of the Krypton after dumping and radiological impact.

7.1.3 Incorporation of Krypton in a solid material

Alternative methods of conditioning the radioactive Krypton shall be developed. The following candidate processes have already been identified :

- implantation by ion discharge in a metal matrix built up by sputtering ;
- adsorption on activated carbon followed by embedding in a low melting metal ;
- encapsulation in molecular sieves.

7.1.4 Incorporation of tritium in a solid material

A screening study, including also laboratory experiments to complete the available knowledge, shall be done in order to assess the potential of various materials for long-term immobilization of tritium. The candidate materials could be : concrete, calcium, sulphate, organic polyners, molecular sieves, metal hydrides, etc.

7.1.5 Immobilization of iodine

An alternative process of conditioning the long-lived radioactive iodine for disposal shall be developed. Preliminary tests indicate the plastic resins and low melting glasses as candidate substances for incorporation of iodine.

8. SEPARATION AND TRANSMUTATION OF ACTINIDES

8 .0 General introduction

The particular problem of the actinides in radioactive wastes is that, owing to their half-lives and alpha-activity, they have to be isolated from the human environment for millions of years. Consequently, the problem of waste disposal would be greatly simplified if actinides could be separated from the other wastes and destroyed, e.g. by being burned as a complementary fuel in nuclear power plants, which would convert them into short-lived or inactive products.

UKAEA and ECN had to carry out (study contract 012-76-WASC) an assessment of the state of the art on the removal of actinides from radioactive wastes followed by their destruction by nuclear processes.

The objectives of this study, which began in May 1976 and will be accomplished at the end of February 1977, are :

- to assess the state of advancement ; this involves surveying the relevant literature, contacting the principal centres in Europe and the USA where work on the subject is performed and making a critical evaluation of the information obtained ;
- to make a preliminary overall assessment (hazards, costs) to the extent that is possible on the basis of the existing knowledge ;
- to locate gaps and to identify uncertainties in the existing knowledge ;
- to describe and assess the future work which is necessary to make a definitive overall assessment.

These objectives shall be pursued in each of the areas of study, which are described in the following paragraphs (8.1 + 8.6).

8.1 Actinide production and their occurrence in waste streams

Starting from work done at the CCR Ispra and elsewhere, complementary calculations will be performed on the basis of a possible EEC programme to 2000 A.D., to be provided by the Commission. Emphasis will be on production in LWRs, LWRs with plutonium recycling, and FBRs, although other reactor types, including those using the thorium cycle, will not be neglected. Significant gaps or uncertainties in the nuclear data required for the calculations will be identified.

Information on the amounts and chemical forms of the various actinides in the different chemical plant streams will be assembled from typical reprocessing centres in the European Community and the USA.

8.2 Separation of actinides

Methods or removal of the different actinides from, especially, the highly-active reprocessing plant wastes will be assessed. Methods which will be considered include solvent extraction, ion exchange precipitation, absorption on solids, fluoride volatility and possibly others. Americium and curium are known to pose particularly challenging problems, owing to their chemical similarity to the lanthanide fission product ; it is envisaged that there will be an initial separation of a combined lanthanide/actinide fraction, followed by isolation of the individual species, and it is recognized that both steps are very difficult. Due consideration will be given to the flowsheets already suggested, though largely untested, for these processes. Treatment of particular chemical forms (suspended solids, colloids, etc.) shall be included in the flowsheets. A tentative to establish the separation factors and the purity required for the separated products will be made.

8.3 Nuclear incineration

Methods of destroying the actinides by nuclear reactions will be considered. Main attention will be given to fast reactors and LWRs in an earlier stage. Significant gaps or uncertainties in the nuclear data required for the calculations will be identified. The relative merits of blending the actinides evenly with the rest of reactor fuel, and of having special actinide fuel elements ("spikes") will be assessed. Some attention will be given to the use of carbide-fuelled reactors and to incineration in a fusion reactor.

8.4 Fuel elements

The problems of fabricating the separated actinides into fuel elements, whether blended with the rest of the fuel or not, will be assessed. The performance of such elements, including their claddings, during the lengthy irradiations likely to be required for nuclear incineration, will be considered.

8.5 Hazard aspects

The relative hazards of the different actinides and their radioactive daughters in the wastes, over long periods of time, will be assessed. The prospective benefits of destruction of the actinides will also be assessed, leading to estimate of the removal factors required for the different actinides species. (It is appreciated, however, that only provisional conclusions can be reached while there are uncertainties about the methods of ultimate disposal, and the pathway to man).

Composition and volumes of waste streams from the conceptual chemical and fuel fabrication plants required for nuclear incineration will be estimated, and, where practicable, their possible treatment will be indicated. Special hazards in handling and transporting recycled actinides, and in operating reactors as nuclear incinerators, will be identified. A preliminary attempt will be made to weigh the hazards due to nuclear incineration against the prospective benefits.

8.6 Logistic and economic assessment

Estimates will be made of the possible scale of nuclear incineration operations in the EEC in 2000 A.D., and especially of the sizes of new plants which would be required. The location of such plants relative to existing reactors and reprocessing plants and the resultant transport problems, will be considered. Timescales will be indicated.

An attempt will be made to give a broad indication of the costs involved.

8.7 Preliminary conclusions

On the basis of the information received some outlines of the general conclusions can be formulated. For the further development of the concept two phases could be distinguished :

- Phase of studies on feasibility and cost/risk/benefit, and choice of a strategy
- Later phase of technological development, leading to a flowsheet and the equipment design for the concepts to be investigated.

It should be considered that the main incentive for actinide separation and recycling will disappear if geological disposal of conditioned wastes can be demonstrated satisfactory for long term.
Attention should be given to the successive recycling of actinide fuels.

8.7.1 Transmutation strategy

The examination of alternative strategies is of first priority to estimate the feasibility and the benefit of the whole concept. Different choices are entailed, such as : reactor designs, burn-ups, special pin or blending into the normal fuel, recycling and so on.

8.7.2 Actinide production

On the basis of the forecasted EEC nuclear programme to 2000 A.D. (1), the JRC Ispra provided new calculations foreseen in para. 8.1 (2).

8.7.3 Chemical separation

Various methods have been tested and possible conceptual flowsheets have been elaborated. Experimental works with real wastes are needed to adapt these chemical techniques to the actual waste.

8.7.4 Nuclear data

The existing data fully demonstrates the feasibility of the concept from the nuclear physics point of view. No improvement is necessary during the first phase .
Needs for reactor safety calculation may be covered by data produced in the frame work of other programmes.

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(1) B. Huber - Estimate of nuclear power growth in the E.C. (Internal document - June 1976)

(2) E. Schmidt - Predictions for HLW to be generated by nuclear power stations in the Member States of E.C. (to be published as EUR report).

9. DEFINITION OF LEGAL, ADMINISTRATIVE AND FINANCIAL FRAMEWORK

The aim of this action is to prevent that the development of nuclear energy after solution of its technical problems is delayed by the absence of a general framework of management in which it should normally be included. The work is conducted directly within a group of experts who assist the Commission and by means of specific contract concluded with consultant firms.

9.1 Legal aspects

A one-year study contract for a comparative investigation of the various national nuclear legislations in the Community has been concluded at the end of 1976 with Environmental Resources Ltd. The study intends to :

- summarize the laws, secondary legislation, regulations, etc... concerning radioactive waste in each of the nine member States of the Community including the incorporation of the relevant provisions of International Law,
- to make an analysis of them,
- to examine their application in practice,
- to compare them and to make a synthetic report, which will include a summary of the main provisions of international conventions, etc...

9.2 Procedures and rules

Customs and rules now in use by the safety authorities as complements, in the radioactive waste field, to the nuclear legislation of general character.

Evolution and trends of legislations.

9.3 Administrative aspects

Administrative aspects ; public or private character of the bodies operating to-day within the Community in the reprocessing, transport, waste storage, waste disposal sectors. Present practice, as far as liability, insurance, etc ... are concerned, for the different sectors.

9.4 Present practice

Present practice concerning the non national wastes ; status of this problem and trends in the different countries of the Community. Necessity of an International convention regarding the management and storage of radioactive waste ; elements of such a convention (strategies; sharing of costs; liability; sites; etc...).

9.5 Financial aspects

Financial aspects; "polluter pays" principle; implementation of such a principle in the member States; role of the governments for financing (disposal; cover of very long term risks, etc.) - present status and trends.

In parallel with these studies, one would try do define what is a radioactive waste and to look at the consequences of such a definition from the legal, administrative and financial points of view.

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