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SITE RESTORATION

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Background

Within the department of Site Restoration, a large variety of activities are carried out to clean up and restore radioactively contaminated sites. The main objective is to participate in delivering a clean and safe environment to the next generations, in the most economical way. To meet this main goal, we develop technologies and procedures for decommissioning, decontamination and waste minimisation, in such a way that the dose uptake and the overall costs are minimised. Our purpose is to protect man and environment from the potential hazards related to contaminated sites and radioactive waste and this starts by preventing and reducing the waste production during the entire lifetime of any nuclear installation i.e. from design through operation and decommissioning.

Objectives

To deliverer a clean and safe site, we:

- perform decommissioning activities in a safe and economical way;
- develop a decommissioning management tool to optimise decommissioning strategies and to identify waste problems;
- prevent the production of radioactive waste where ever possible, and contribute in reducing the volumes of nuclear waste;
- manage waste in a safe and economical way according to the legal rules leading to the reduction of the impact of the waste to man and environment;
- provide our expertise in decommissioning and waste minimisation to international institutions, regulatory bodies and companies;
- develop, assess and apply new and existing treatment/conditioning processes (up to the demonstration phase) minimising the risks, the volumes and the costs of nuclear waste;
- provide for the new treatment/conditioning processes reliable data and models as well as integrated overall approaches to the design engineers with a view to determine the final plant characteristics.

Programme

To reach the main objective to deliver a clean and safe site and to convert experience into exploitable expertise, our activities can be divided into three main categories:

- activities carried out to clean up SCK•CEN's own contaminated sites;
- consulting activities carried out for third parties using own expertise in cleaning up contaminated sites;
- development of new processes to continuously expand and improve SCK•CEN's experience and expertise.
- **SCK•CEN' own needs** required to carry out the following works:
  - clean-up activities;
  - waste minimisation activities;
  - activities related to the management of decommissioning projects.
- **SCK•CEN's expertise** was asked by **external parties** for:
  - the set up of a remote tooling for dismantling the core shroud of a commercial BWR in Sweden (Forsmark NPP, in collaboration with ABB-ATOM);
  - the set up of the physical and radiological inventory as well as the decommissioning plan for the Thetis reactor (University of Ghent - Belgium);
  - the management of the waste stored at the Interim Storage Facility situated at the Josef Stephans Institute (Slovenian radioactive waste management agency: RAO-Agency);
  - the review of the methodology used by Nuclear Energy Corporated South Africa to assess the liabilities coming from the former Atomic Energy Corporated (AEC) (Department of Art Culture Science and Technology (DACST), South Africa);
  - the organisation and the management of the decommissioning project of Sallaspils (Lettland) as well as the improvement of the decontamination (International Atomic Energy Agency);
  - the organisation of educational and training courses.
- We launched research related to **the development of new processes** or continued to increase our knowledge and expertise. It concerns:
  - the treatment of tritiated methanol;
  - the separation of uranium and plutonium from solutions of fresh and spent fuel;

- the treatment of alkali metals;
- the electrochemical mediated oxidation of organic material by Ag^{2+}
- the selective extraction of Co out of effluents.

Achievements

SCK•CEN's own needs

Clean-up activities

We focused the activities carried out to clean-up SCK•CEN's own contaminated sites on four installations, i.e. the pressurised water reactor (BR3-PWR), the materials testing reactor BR2 (MTR-BR2), the Laboratory of High and Medium level Activity (LHMA) and the Chemistry Building (SCH). We also made progress in the application of the defined solutions concerning the back-end of the fuel cycle of the BR3 and the BR2 reactors.

Decommissioning of the BR3 (PWR)

The main decommissioning activities performed included the dismantling of the reactor pressure vessel, the primary and auxiliary loops under the operating deck, the turbo-alternator and the removal of the asbestos around the auxiliary loops.

The *BR3 reactor pressure vessel* (the figure below shows its installation at the beginning of the 60's) is activated up to 7.2 MBq/kg in the stainless steel cladding at mid-plane. Therefore, the reactor pressure vessel has to be dismantled by remote controlled tools. We chose for the under water dismantling of the reactor pressure vessel, since we used this cutting strategy previously at the BR3 for the dismantling of the two sets of reactor internals. The detailed study showed that the reactor pressure vessel dismantling was feasible reusing the same supporting and cutting equipment. This approach also allowed us to reduce the operation duration, and consequently the dose uptake respecting the ALARA (As Low As Reasonably Achievable) principles.

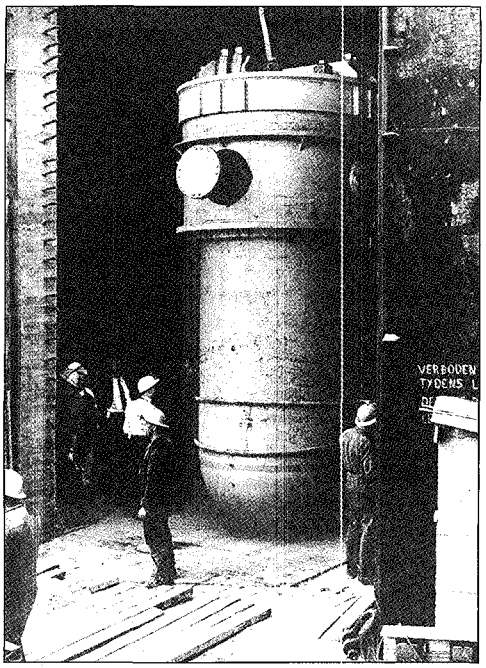
Nevertheless, the reactor pressure vessel dismantling presents significant differences in comparison with the dismantling of internals. The reactor pressure vessel has a larger diameter (1.7 m compared to 1.2 m) and is much thicker (114 mm compared to a mean thickness for the internals of about 30 mm). The reactor pressure vessel has a higher mass (about 26 tons compared to 5 tons) and is also made of two different materials (carbon steel (cladding) and stainless steel instead of stainless steel only for the internals). Therefore, additional clamping devices were required to compensate for the additional generated cutting forces. Other cutting parameters associated with other saw blade types and geometry's were also needed. The new cutting parameters were selected based on the intensive cold tests we performed in 1999.

The selected dismantling strategy involves firstly the underwater removal of the entire vessel from its original position (i.e. under the bottom of the refuelling pool) to the refuelling pool.

This approach presents some important advantages:

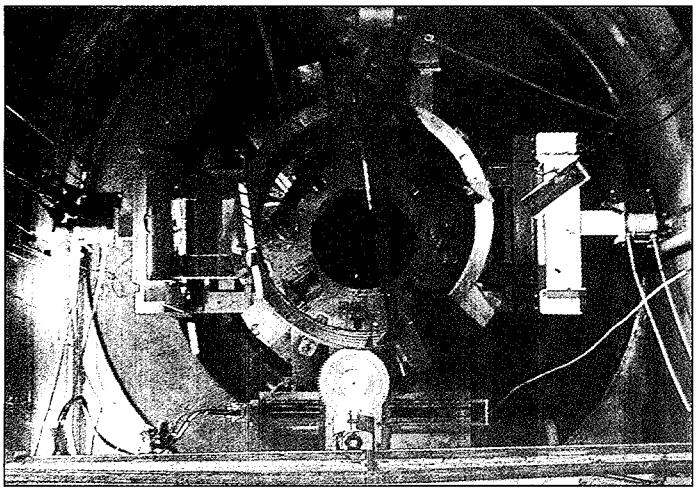
- a thick layer of water above the high active parts of the reactor pressure vessel ensures shielding allowing us to respect the strictest radioprotection requirements;
- additional accessibility for tools at the outside of the reactor pressure vessel wall was essential to remove the surrounding thermal insulation and its maintaining shroud;
- the risk of spreading contamination during the cutting process and manipulation is reduced to its minimum. Indeed, at BR3, in conjunction with underwater segmentation, mechanical cutting is promoted. Consequently, the chips produced fall on the bottom of the refuelling pool and can be collected without any difficulty using an underwater vacuum cleaner.

At the beginning of 2000, after a year of intensive preparation, the reactor pressure vessel was hanging at its crane, ready to be segmented. After the removal



BR3 reactor pressure vessel installation at the beginning of the 60's.

of the reactor pressure vessel insulation and its shells, the reactor pressure vessel bottom was cut first. The vertical cylindrical component of the reactor pressure vessel is then cut into 9 rings using a milling cutter machine (see picture below). Afterwards, we cut each ring into 12 segments using a band saw machine (cutting vertically).



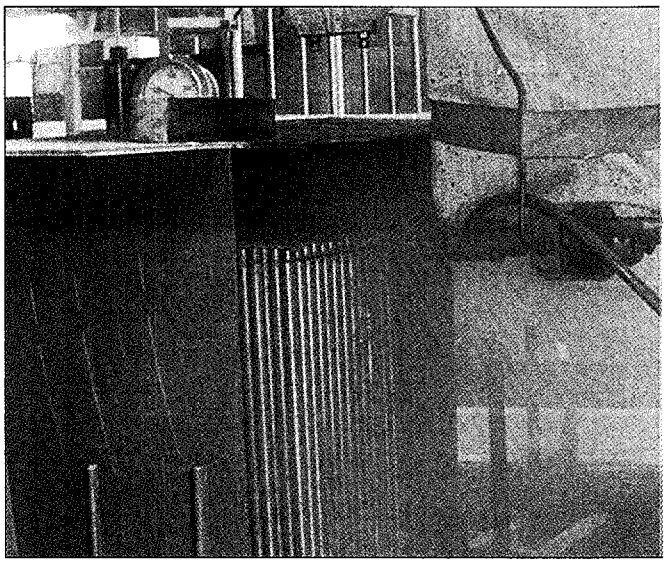
Reactor pressure vessel horizontal cutting with anti-vibrations systems.

The cutting levels and the ring segmentation were optimised in order to minimise the conditioning and disposal costs, which increase strongly with the waste categories. We defined the horizontal cut levels based on contact dose rates, on activity measurements of samples extracted from the vessel in 1995 and on activity measurements carried out on internals swarfs (correlated by neutronic calculation). The predicted values were in good agreement with the real ones for about 80% of the rings. All the waste produced by the dismantling of the reactor pressure vessel was transferred to Belgoprocess for conditioning and interim storage awaiting for the final disposal.

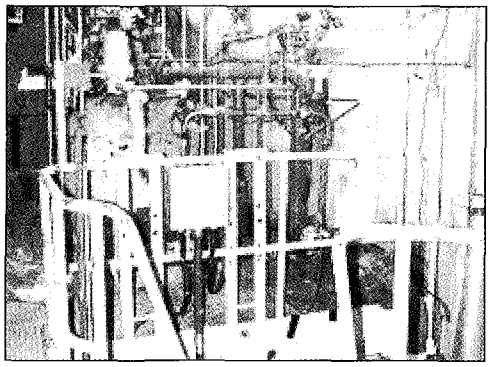
The *large components of the primary loops* i.e. the steam generator, the pressuriser, the primary pump housing, the neutron shield tank (surrounding the already removed reactor pressure vessel) and the reactor pressure vessel bottom and cover head can be considered as complex components to be dismantled. They require a light and handy tool to reach the difficult area and to follow exactly actual 3D surfaces.

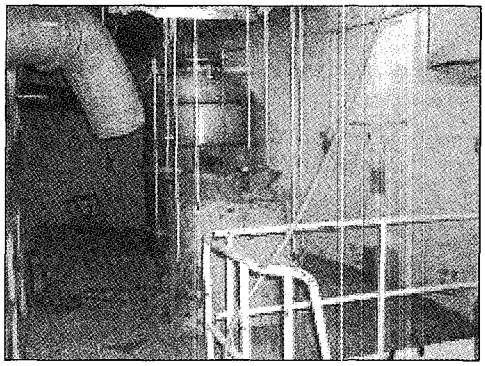
We chose high-pressure water jet cutting (HPWJC) with abrasives as dismantling technique. To handle it, we ordered a hydraulic telemanipulated arm. Once again, we will use the refuelling pool as cutting workshop. During 2000, we studied the telemanipulated equipment in detail and then we built it. On the other hand, we tested the cutting techniques on realistic mock-ups to optimise the entire operation. The cutting speed should be as high as possible to reduce the operation duration, and consequently the dose uptake. We selected an average cutting speed of about 20 mm/minute (a wide range of speeds is required depending mainly of the thickness of the material to be cut). The abrasive flow rate has to be kept as low as possible to minimise the production of secondary waste. We chose an abrasive flow rate of 250 g/minute.

The removal of the steam generator and the pressuriser requires the dismantling of the *primary loop and auxiliary under the operating deck*. This decommissioning concerns a large amount of electric cables and cable runs, pipe supports, valves, pumps and electric motors. The complete dismantling strategy was set up after a detailed ALARA study. This study was conducted in collaboration with the Health Physics and Safety department and the SCK•CEN's software VISIPLAN. We removed easily accessible hot spots to reduce the ambient dose rate. Then we carried out a systematic dismantling in two phases. We performed first the in-situ cutting, using mechanical techniques where possible. The dismantled components were subsequently segmented in smaller parts, either in situ with the band saw on a frame or in a ventilated workshop using thermal techniques.



Cutting of a set of small tubes simulating the steam generator bundle.





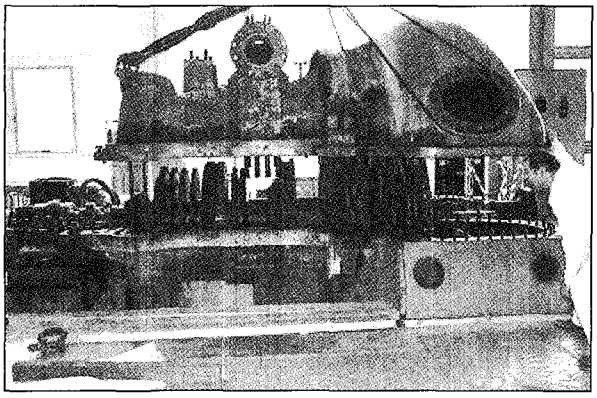
Dismantling of valves and pipes of the pressuriser and of the Spray Tank.

About 20 tons of material were dismantled and will be removed after their respective waste minimisation treatment. The scaffolds, necessary for the dismantling of the steam generator and the large pipes (primary loop), as well as a monorail with pneumatic hoists for the evacuation of heavy pieces are both installed.

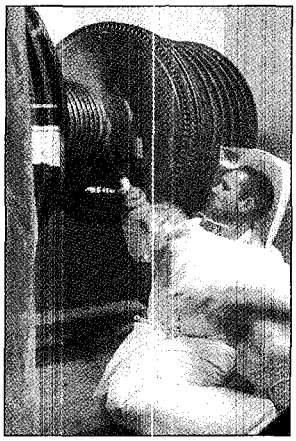
The *turbo-alternator group* was situated in the machine hall of BR3 and comprised the turbine and the alternator. The turbine was made of two major parts (high pressure and low pressure), without intermediate bearing. It was coupled to the main and auxiliary alternators, situated at the same frame and with an effective power of 11.5 MWe.

The main and auxiliary alternators were not contaminated and could thus be free released. However, the turbine and connected loops were considered as suspect as a result of leakage's in the secondary loop during the operating period (at the level of the steam generator tube bundle). Before dismantling the turbine, we removed asbestos and glass wool. All of this waste could be removed as classic non radioactive asbestos and has been free released. The turbine dismantling operations were carried out as described in the intervention and maintenance procedures during the operation period of the plant. To avoid the construction of a confined/ventilated area, we used manual disassembly and cold techniques (due to the suspected contamination). For several cuts however, we used an oxyacetylene torch. The Health Physics and Safety department took samples at the cutting places to check that the contamination is limited and confined sufficiently. The turbine shaft was decontaminated with dry ice (CO_2) and wet sand blasting. The turbine shaft was free released .

In the BR3 an external and certified company has carried out the asbestos removal works. To reduce the total waste volume, a reconditioning of the waste is envisaged. These circuits are now ready for further dismantling.



Turbine



Decontamination of the shaft with dry ice in a closed/ ventilated area: operator in ventilated suite.

Cleaning activities in other SCK•CEN facilities

In the BR2 reactor, we dismantled an old experiment and its bunker. Almost 21 tons of metal components and 100 tons of concrete blocks were dismantled and controlled on radioactivity. Only 3 tons of concrete blocks had to be decontaminated to reach the free release level.

The decommissioning activities at LHMA mainly concerned the dismantling and the decontamination of hot cells. We paid special attention to the hot-cell 41 where spent fuel has been dissolved. The final aim of the decommissioning of the hot-cell 41 is to assure its reuse after sufficient decontamination during a refurbishment period. First, we disassembled the easily removable internals, followed by a first decontamination of the hot cell 41 to minimise the dose to the workers during the further decommissioning intervention (ALARA principle). The next decommissioning steps will involve the dismantling of the movable bridge and a lead-shielding chamber followed by a final decontamination.

Management of spent fuel

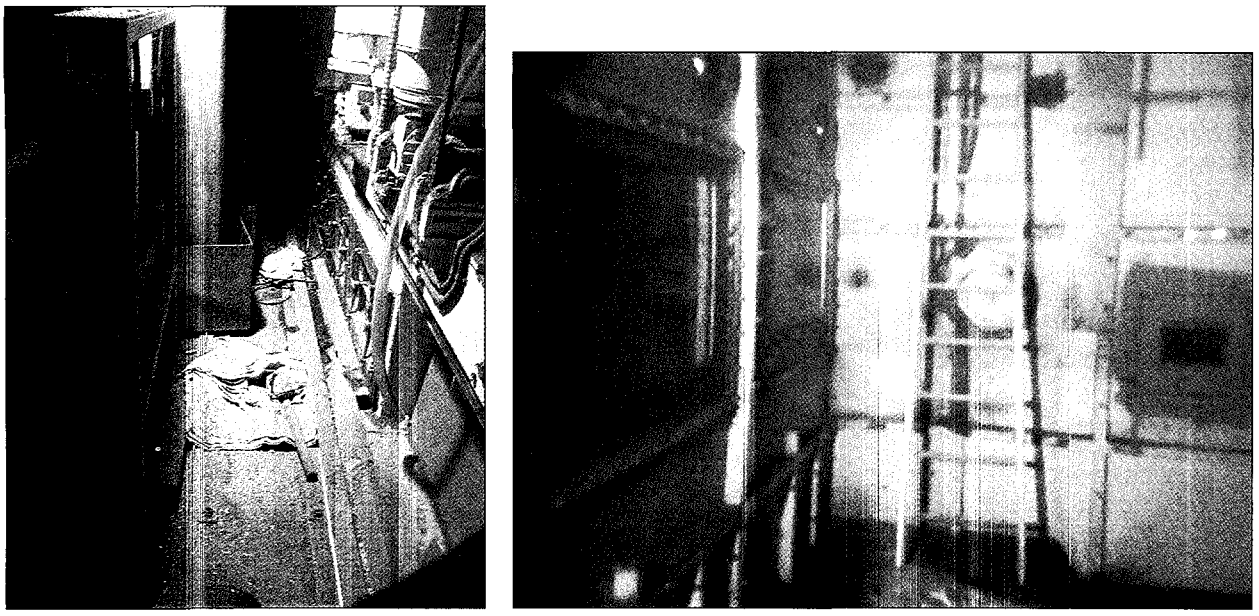
Further clean-up activities involve the transport to La Hague for reprocessing 340 spent fuel elements of the BR2 reactor. Most of the work related to the BR3-spent fuel preparation concerning the consolidation of fuel assemblies and their "Wet-Sipping" was completed.

On March 28, 2000 the first CASTOR BR3® cask (cask for interim storage of spent fuel) was cast. This event was the first step in the fabrication process. It will take 11 months to finish the first CASTOR BR3®, and afterwards, the other 7 containers will be produced in a period of 4 months. We submitted the documents for the transport licence to the authorities in March 1999. The documents for the storage licence were submitted during the month of September 2000. Loading of the CASTOR BR3® casks is foreseen in 2001, if we obtained all necessary licences.

As a result of SCK•CEN's R&D programs in the characterisation of Low Enriched Uranium (LEU) and Mixed OXide (MOX) spent fuel, considerable amounts of damaged fuel pins and segments are in temporary storage facilities waiting for a final solution. During 2000, the repackaging of LWR-type fuel rod remnants (LEU and MOX) for long term intermediate dry storage were almost completed. The remnants were retrieved from their temporary storage facility and conditioned into welded canisters, provided with an inner Zr-tubing basket. Those canisters will later be loaded into CASTOR BR3® casks for further long-term intermediate storage (50 years). In parallel with the conditioning, data were collected to calculate the nuclide inventory of the canisters.

Waste minimisation

The dismantling activities presented above produce a large variety of materials ranging from electric cables up to stainless steel reservoirs. Since we want to minimise the amount of radioactive waste, we are using two main alternative evacuation routes, i.e. the



Dismantling of the spent fuel cell 41: before and after intervention.

recycling route (the reuse of the materials inside the nuclear world) and the clearance route (unconditional reuse of the materials for the industrial world). A decontamination process can precede the clearance route.

The crucial point in the waste minimisation process is the sorting of the materials. Specifications are established to help the operator in his choice. The sorting of the materials leads to the creation of batches i.e. materials that follow the same evacuation route. An interactive database allows the follow up of the materials from the creation of the batch up to its final evacuation.

The recycling of low contaminated metals uses a radioactive foundry. The product, i.e. ingots, is used for the fabrication of shielding blocks or for the fabrication of waste containers. Up to now, 64 t of mild and stainless steels have been sent to GTS-Duratek (USA).

Some materials are either very low contaminated, very difficult to measure or not homogeneously contaminated. For these materials, it is advantageous to send them to a nuclear foundry to reach after melting the clearance level. Melting further decontaminates the metals by volatilisation (e.g. ^{137}Cs) or by transfer of the radionuclides to the slag. It allows also an accurate determination of the radionuclides content thanks to the homogeneity of the metal melt. We performed in 2000, a first transport of 18 t of metals to the Studsvik facility in Sweden. This transport comprised secondary reheaters with copper tubes, a massive carbon steel component and carbon steel and stainless parts in 200 l drums. All the ingots produced were unconditionally cleared. The secondary waste, representing only 5.9 % of the primary weight, will be sent back to Belgium for conditioning.

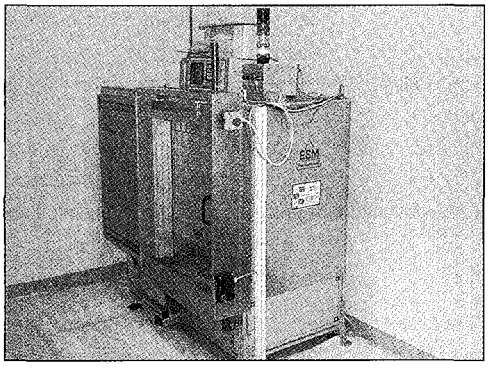
The clearance procedure for metallic materials was, up to now, limited to the clearance of materials with simple geometry of which 100% of the surface was measured by hand held β monitors. We developed with the Health Physics and Safety department a procedure for the clearance of materials with complex geometry. This procedure comprises now two steps:

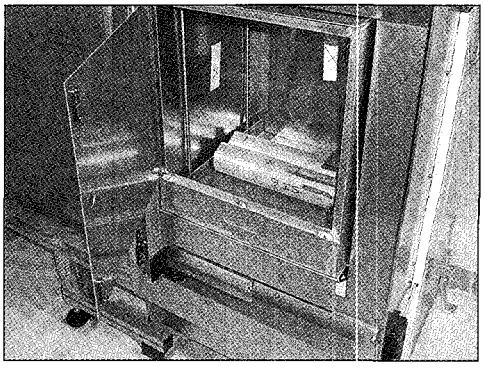
- a first 4π measurement: measurement by small batches of materials (1/10 of a 200 l drum) by gross gamma counting with scintillation detectors. This measurement allows detecting "hot spots" of contaminated materials in a batch of materials;
- a clearance measurement of a 200 l drum with the existing gamma spectrometer with High Purity Germanium (HPGe) detectors. This measurement verifies that the nuclide specific clearance criteria are respected.

A new measuring equipment, the FHT 3035 clearance monitor developed by ESM (Erlangen, Germany) has been installed at BR3, calibrated by the Health Physics and Safety department and approved by the authority. This equipment comprises a 200 l closed shielded chamber equipped with 6 plastic detectors. The apparatus uses a new technology, the CCM system (Cobalt Coincidence Method) which allows to distinct somewhat between ^{137}Cs and ^{60}Co, which are the main gamma emitters at BR3. The mass of a typical batch measured reaches almost 20 kg.

This equipment is now used in routine at BR3 for the measurement of dismantled components; the main limitation is the maximum size of the component (max 700 mm long) and the maximum mass of one individual part (60 kg).

To reach the clearance level, decontamination techniques can be used. For metals, we mainly use manual washing or cleaning in an ultrasonic rinsing bath, a wet abrasive decontamination (ZOE cabinet) and a hard chemical decontamination based on a cerium process, called MEDOC (MEtal Decontamination by Oxidation with Cerium).

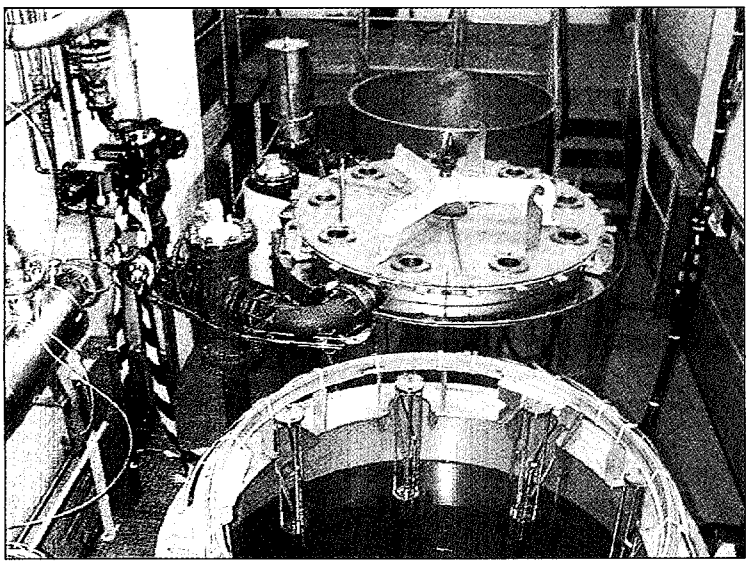




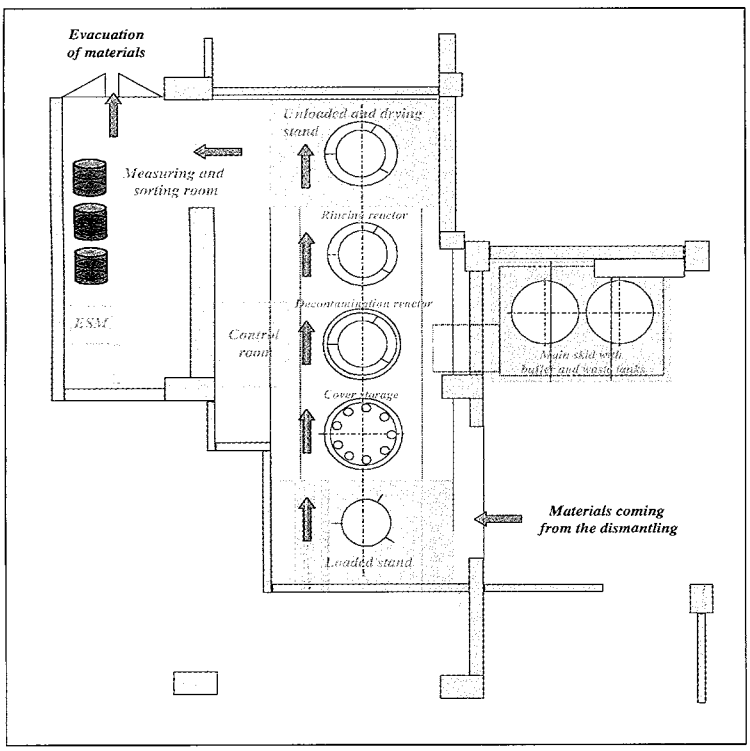
The new ESM-FHT 3035 clearance monitor installed in a low background area.

The manual washing or cleaning in an ultrasonic rinsing bath is mainly used for components slightly contaminated on external surfaces (e.g.: demineralised water piping, structural pieces, instrumentation boxes ...). During 2000, we treated about 5.7 tons in this manner.

The ZOE cabinet allowing the decontamination by a wet abrasive process is mainly used for rusted or painted components with simple geometry in which the contamination is fixed in the oxide layer or in the paint (structural equipment, beams ...). ZOE is used in routine for the treatment of components up to 3 t and 3 m long maximum. During 2000, ZOE treated 6 tons.



View of the MEDOC decontamination installation after installation at BR.3



Material stream in the MEDOC workshop.

The hard chemical decontamination process, the so-called MEDOC, is based on the use of cerium IV as strong oxidant in sulphuric acid with continuous regeneration of the cerium using ozone. An industrial installation has been designed and constructed in close collaboration with Framatome (France). This installation started to operate in September 1999 for the treatment of the metallic parts arising from the dismantling of the BR3 reactor.

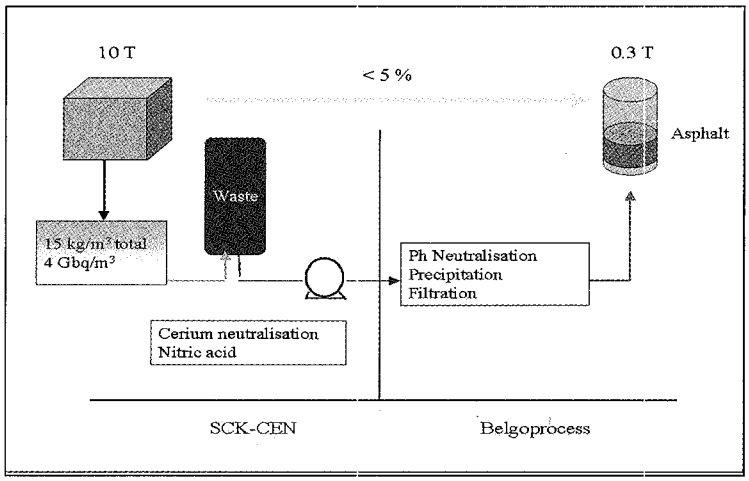
The decontamination process can be summarised as follows. The decontamination loop, loaded with batches of 0,5 to 1 ton of contaminated materials, comprises a decontamination reactor, a buffer tank, a circulating loop and a static mixer where the ozone is injected. Ultrasonic transducers surround the basket to promote the decontamination process. The decontamination is performed at 80°C in a sulphuric acid solution loaded with ceric sulphate. Contaminated materials are attacked with an average corrosion rate of about 2.5 $\mu m.h^{-1}$. This corrosion rate gives an idea of the necessary treatment time. In order to remove 5 µm on low contaminated material to 15 µm for high-contaminated material, the treatment time is situated between two to six hours. The continuous regeneration process with ozone-laden oxygen compensates the consumption of the ceric ions by reaction with the metals. The regeneration efficiency being over 60% allows us to maintain the cerium IV concentration constant as well as the corrosion rate. After decontamination, treated materials are removed from the decontamination reactor and transported inside a rinsing reactor. The rinsing is then performed in a filtered closed loop with ultrasonic transducers surrounding the basket to promote the rinsing effect. When the rinsing is finished, the basket is placed on a drip tray to allow drying of the components. The components are then manually transferred inside a clean container to be sent to the measuring station. Up to now, 77% of these decontaminated materials have an average residual activity close to 0.05 $Bq.g^{-1}$ of ^{60}Co and follow the free release route; 23% have a residual activity lower than 1 $Bq.g^{-1}$ and follow the melting route for free release. The pictures below give a view of the MEDOC installation and the material stream into the MEDOC installation.

Since the contamination level and the salt content of the decontamination solution continuously increase,

the solution must be evacuated as radioactive waste after a certain time and/or a certain number of treated batches. The liquid waste is then transported to the Belgoprocess liquid waste treatment unit for final conditioning. The conditioning method developed by Belgoprocess comprises a neutralisation/flocculation step followed by bituminisation of the obtained sludge. The overall process starting from contaminated materials to final conditioning guarantees a total volume-reduction factor close to 95 %.

The main measuring equipment is the FHT clearance monitor, which is installed in the vicinity of the decontamination installations. It allows us to take immediate corrective actions in case of residual contamination.

Within one year, we treated about 14 tons of stainless steel in the MEDOC installation. The license of the patented MEDOC process has been granted to the Framatome company for all countries except the Benelux.



Secondary waste removal and treatment routes for the MEDOC decontamination installation.

Management of decommissioning projects

Our Decommissioning Management Tool (DEMATO) is now adapted to use also the structure of the inventory of nuclear installations as defined in 1999 by the Belgian Agency for Radioactive Waste and Enriched Fissile Materials (ONDRAF/NIRAS: Inventory of Nuclear liabilities). The upgraded DEMATO modules assessing the liabilities of nuclear installations and fissile material will be fully tested in the near future. We will compare the results with those obtained by the previous release of DEMATO and by the return of experience from the ongoing decommissioning projects.

We modelled the structure materials of the BR1 reactor (i.e. the graphite moderator and the concrete bunker) and of the critical VENUS assembly (i.e. the stainless steel vessel and the concrete shaft) to gain information on the activation products. The results of the modelling were incorporated in the inventory database and allowed us to reassess the decommissioning costs of these facilities.

External consultancy and services

Set-up of a remote tooling for dismantling the core shroud of a commercial BWR in Sweden

For maintenance purposes, the nuclear power plant Forsmark in Sweden had to replace its core shroud. The old core shroud being stored in the refuelling pool, had to be dismantled, while the plant was running at full power. The plant owner preferred mechanical cutting above any other method, to minimise the possibility to pollute the reactor pool. We used our expertise in underwater reactor internals dismantling to assist the industrial partner (ABB-ATOM Sweden) in designing the tool for dismantling the core shroud.

Set-up of the initial decommissioning plan of the THETIS reactor (University of Ghent, Belgium)

According to the Belgian national regulations, every nuclear installation has to dispose of an inventory of its own nuclear liability and of a decommissioning plan. The University of Ghent chose SCK•CEN to set up the physical and radiological inventory and to draw up the initial decommissioning plan for their THETIS reactor. We paid special attention to the modelling of the activation of the graphite reflector and the heavy concrete of the reactor shaft to specify the decommissioning strategy and the waste management route. The initial decommissioning plan, conform to Belgian and international (International Atomic Energy Agency) recommendations, was approved by the University of Ghent. SCK•CEN's own developed software tool (DEMATO) was used for calculating the total decommissioning cost and setting up the financing scheme.

RAO-Agency (Brinje, Slovenia)

SCK•CEN made an evaluation of the storage conditions of the historical waste stored at the Interim Storage Facility situated at the Josef Stephans Institute (Brinje, Slovenia). We made recommendations on the improvement of the safety of the storage facility and on the rearrangement of the historical waste allowing to define adequate treatment and conditioning routes. Further, we recommended on waste record keeping and on acceptance criteria of low and intermediate level waste resulting from medical applications, research and the industry.

Expert missions

- We participated in the review team constituted by the Department of Arts, Culture, Science and Technology (DACST, South Africa) on behalf of the Department of Minerals and Energy (DME), to undertake a *"review of the procedures for assessing the decommissioning and waste management liabilities arising out of the past activities of the former Atomic Energy Corporation (AEC)*";
- We were appointed as international expert in an ad-hoc Committee of the US National Academies of Science, on the "Long term research needs on D&D at the DOE". We also participated, as Belgian representative, to the expert Committee set up by the European Commission for advising on its projects of decommissioning, clean-up and waste management of the nuclear liabilities in its Joint Research Centre sites, with a particular emphasis on the Ispra site;
- In the framework of IAEA bilateral co-operation agreement, we were also involved in several expert missions in Latvia.

Valorisation

We participated in several calls for tender together with industrial partners. Together with Framatome, we are developing and testing, at lab scale, different methods and processes to clean-up the Dounreay spent fuel pond.

We will soon be involved in the preparation of the decommissioning plan of Kozloduy in Bulgaria (with Belgatom) and the set-up of a Methodology for Decommissioning Costs (with Framatome, EDF and VUJE – local contractor).

Our expertise in decommissioning was brought to the attention of the Central Research Institute of Electric Power Industry (CRIEPI) in Japan which is in charge of evaluating the environmental impact assessment of decommissioning nuclear power plants in Japan.

We sold the license for the MEDOC process to Framatome and the VISIPLAN ALARA planning tool to Framatome and Tractebel.

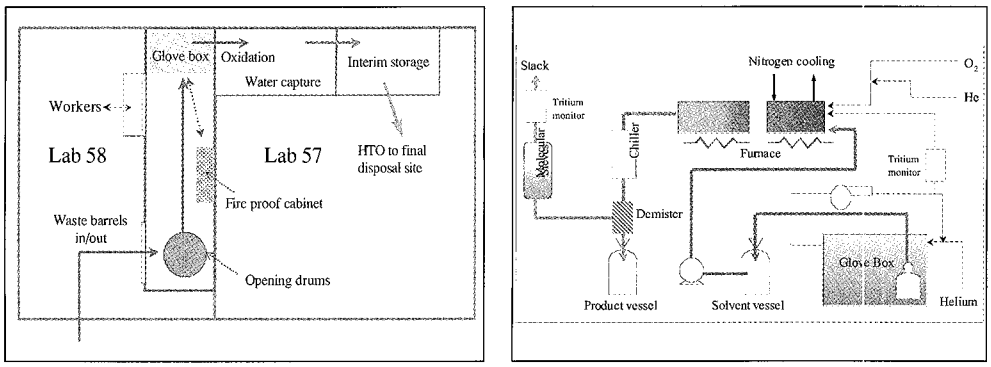
We trained a decommissioning engineer from the SOGIN company (Italy) who is in charge of the decommissioning of all nuclear power plants in Italy and we gave lectures at the IAEA interregional training course on decommissioning small nuclear installations (Argonne, USA).

We provided also training and courses for universities (Ulg, Liège, Belgium) and national institutes e.g. Nuclear Power Institute of China and Ente Nazionale per l'Energia Elettrica (ENEL, Italy).

Development of new processes

Processes related to tritium

SCK•CEN treats tritiated waste, mainly methanol, produced by the pharmaceutical industry. We intend to convert this tritiated waste in tritiated water, which can be conditioned and stored, and in carbon dioxide, which can be discharged. The requirement for negligible environmental emission drives the need for high destruction efficiencies and efficient product-water recovery. At SCK•CEN, we realised an appropriate infrastructure (see figure below) which allows a controlled and safe handling of this volatile, highly flammable, toxic and highly radioactive material. The installation comprises a process cell equipped for unpacking and sampling the waste, a second cell which houses the oxidation system and its associated water collection system and a third cell equipped for the temporary storage of the tritiated water. A combination of direct combustion and catalytic after-oxidation, developed and tested by Ontario Power Technologies Canada will ensure the very high conversion rates that are required for environmental reasons. During 2000 it was integrated in our infrastructure and cold tests allowed us to optimise working conditions. Condensation and adsorption on molecular sieves will trap the tritiated water. At the end of 2000 the regulatory body granted the exploitation licence. Soon thereafter we started unpacking and sampling of the waste.



Treatment of tritiated waste: overview of the tritium route and schematic design of the process.

We started opening of all secondary overpacks in a ventilated process cell, using air-supplied suits and preventing sparks. This operation will end during the first trimester of 2001. During this operation, we continuously monitor for tritium, methanol and radiolytically produced hydrogen and decontaminate the drums and overpacks. For removing all primary overpacks and sampling waste bottles, we use a glove box, the inert atmosphere of which is decontaminated in the processing installation. Sampling allows us to check the chemical composition, the specific tritium activity and the absence of other radionuclides. After sampling, the waste bottles are stored in a fireproof cabinet. Up to now unpacking of the drums has posed no significant problems and tritium discharges have been well below the authorised limits. We plan to finalise unpacking in the beginning of 2001. After the exact chemical composition of the waste has been determined, the actual oxidation will start, with optimal parameter settings depending on the composition. The resulting water will be stored at SCK•CEN, awaiting final conditioning and disposal at Belgoprocess.

In the framework of the ELEX process for water detritiation, SCK•CEN developed and tested a suitable hydrophobic catalyst, which does not loose its activity in the presence of liquid water. Our catalyst contains platinum on a carrier of charcoal incorporated in a polytetrafluoroethylene matrix. Mixed with water wettable packing material in a volumetric ratio of 3:1, this catalyst allowed us to realise overall tritium exchange rate constants up to 100 $mol.s^{-1}.m^{-3}$ at 40°C. Linked with fusion research and tritium tests in Europe and elsewhere in the world, we experienced a renewed interest in liquid phase catalytic exchange and the possible use of SCK•CEN's proprietary catalyst for water detritiation. With the help of ZeTek Power bvba, SCK•CEN prepared and sold in 2000 several samples of its hydrophobic catalyst.

Processes related to nuclear fuel

In the framework of the development of an advanced wet-reprocessing process to separate uranium, plutonium and minor actinides from a nitric acid solution of spent nuclear fuel, the Japanese Institute of Research and Innovation (IRI) is manufacturing a novel anion exchanger (AR-01) and several extraction resins immobilised in porous silica particles. In comparison with the Purex process, the new process would exclude or limit the use of organic solvents, produce less waste and be less expensive because of compactness and simplicity. The new process is based on ion exchange in combination with electrolytic reduction and extraction chromatography. SCK•CEN was asked by IRI, to test their new anion exchanger AR-01. In nitric acid solution, uranium and plutonium exhibit distinct retention on an anion exchanger as they form negatively charged complexes, while fission products mostly exist as cations and show no or very weak retention. After promising results with simulated spent fuel solutions in 1999, we studied in 2000 the separation of uranium and plutonium from a solution of real spent fuel. The experiments at SCK•CEN were carried out in collaboration with Nuclear Chemistry and Services and Reactor Materials Research department. We cut a small piece of LWR fuel with a burnup of 55000 MWd/tHM, dissolved it in 8 M nitric acid and adjusted the concentration to 0.5 M uranium and 6 M nitric acid. 40 cm^3 of the spent fuel solution and several eluent and rinse solutions were successively supplied to a glass column packed with AR-01 and operated at 296 K and at a superficial flow rate of 3.8 $m.h^{-1}$. Element or isotope concentrations in the feed and in the collected effluent fractions were determined using inductively coupled plasma - mass spectrometry and alpha or gamma spectrometry.

The results indicated that most fission products such as cesium, strontium, molybdenum, rhodium and the

lanthanides as well as the minor actinides americium and curium were nearly not retained from a 6 M nitric acid solution and they were satisfactorily separated from uranium. Changing the eluent to 1 M nitric acid eluted the retained uranium. Zirconium, niobium and ruthenium exhibited weak retention and were partly mixed with the effluent uranium fraction. Plutonium was completely retained, but by using 1 M formic acid as the eluent, it was successfully and sharply separated from uranium and fission products. According to our complementary batch study of Pu(IV) distribution coefficients and UV-spectra the elution of plutonium is probably based on the formation of a non anionic plutonium formate complex. Neptunium showed a complicated elution behaviour due to its different oxidation states. Np(V) and Np(VI) were mostly mixed with the uranium fraction, while Np(IV) was found in the plutonium fraction. Palladium and technetium were strongly fixed on the anion exchanger and their elution, if necessary, needs further investigation.

The incomplete separation between uranium and some fission products is considered to be due to the relatively low retention ability of U(VI) on the anion exchanger. To enhance the separation between uranium and these fission products, IRI proposes to electrolytically reduce uranyl (UO_2^{2+}) to uranous ion (U^{4+}), which shows a considerably stronger retention than UO_2^{2+} in nitric acid solution. In 2001 SCK•CEN will study this electrolytic reduction and the further purification of uranium on AR-01.

To maximise recycling of nuclear material and to minimise waste production, SCK•CEN is recovering fissile material from fresh-fuel solutions for BELGONUCLEAIRE and other clients. The installation is enclosed in three interconnected glove boxes, which contain respectively the precipitation and filtration section, the drying oven and the calcination furnace. After adding ammonia to the uranium and plutonium containing solutions at 60 °C, we filter and rinse the precipitate and dry it at 200 °C under inert atmosphere. Afterwards we calcine it at 600 °C in the presence of hydrogen. The fully characterised product returns to the client. During 2000 about 125 dm^3 have been treated and about 7.5 kg uranium and 0.5 kg plutonium have been recovered. Because of a necessary revision of the precipitation section, other obligations and a lack of manpower, the recovery of fissile material has to cope with an important backlog, necessitating an extension of our storage capacity for untreated solutions.

The oxidation of alkali metals

SCK•CEN decided, on the basis of an elaborated business plan, to continue developing a novel and patented process for the oxidation of radioactive alkali metals. The main objective of this process, called SANDS (for SAfer Na Destruction System), is to develop a safe method for the handling of contaminated alkali liquid metals (Na, K) leading to a final product (sand like) directly ready for waste conditioning (either by cementation or by vitrification). The process (now patented) is based on the reaction of the liquid metal with CO_2 in a controlled fluidised bed.

On a theoretical level, we further developed two heat transfer models for the fluidised bed reactor. The first model includes heat conduction and convection. The second model combines the ideally mixed conditions with the plug flow regime. Adequate experimental campaigns for validating the models will require time, due to the size of the experimental apparatus and to safety related constraints.

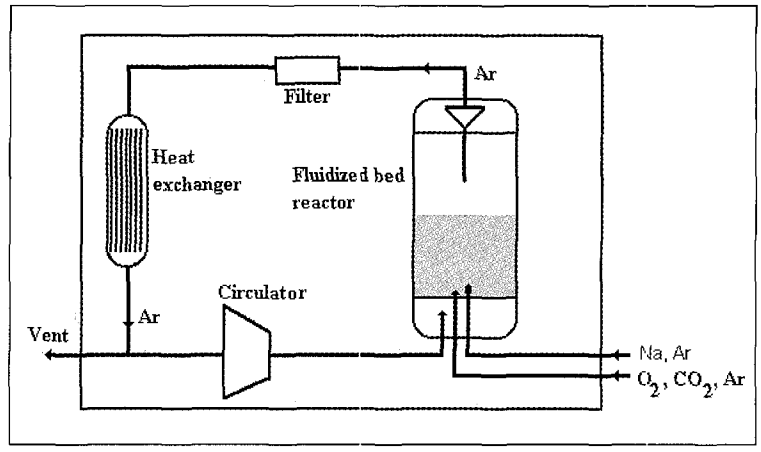
On the experimental level, we carried out a first experiment during which 1 kg of sodium was injected into the fluidised bed reactor. The figure below shows the schematic design of the treatment installation. The injection line worked well. The system reached its fluidisation regime normally. However, the conversion of sodium was very low due to an internal leak of the cooling fluid into the reactor vessel. This resulted in a passivation of the sodium droplets. In these conditions, the initial temperature (403-413 K) was too low to ignite the sodium. We reviewed the installation, always keeping safety into account. We replaced the internal joining by a weld. The procurement of adequate materials in view of increasing the pre-heating temperature is finished. Further, using the injected sodium in the sand matrix, we carried out laboratory experiments, using 5-15 vol. % CO_2 and 20-30 vol. % O_2 in Ar. We demonstrated that peroxides can be formed. Next, an initial temperature of 543 K would better guarantee ignition in most cases. In these conditions, both the carbonation and the oxidation can be complete. The experiments that were carried out made us realise that we might work in two stages: first the oxidation, next the carbonation. This is because traces of water vapour enhance considerably the kinetics of the carbonation step while we reject water during the oxidation stage for safety reasons. We plan to carry out the next experiment with at least 1 kg of sodium at the beginning of 2001 after approval of the related safety report.

The oxidation of radioactive organic waste through Ag^{2+}

SCK•CEN decided, on the basis of an elaborated business plan, to continue developing a modified process for the oxidation of organic material using Ag^{2+}, for which a patent application has been filed.

On the theoretical level, an experimental campaign was carried out to assess a model describing the anodic selectivity when oxidising formic acid. Data treatment through fittings and statistical analysis is still going on.

On the experimental level, we designed several experimental devices with the aim to study membrane characteristics. Further we investigated the possible replacement of nitric acid by sulphuric acid in the catholyte. This would lead to the production of hydrogen instead of HNO_2 and/or NOx. The questions raised during this investigation are similar to those we are facing with nitric acid on both sides of the membranes. Further, we examined several different methods for the online monitoring of traces of H_2O_2 in concentrated HNO_3 solutions. These methods are based on the colorimetric determination of the complex formed between H_2O_2 and Ti^{4+}. A homemade online colorimeter (combining an adequate blue LED and a phototransistor) was used for these measurements. These methods do not allow coping with the low dead time of the targeted control loop (less than one minute). To ensure an integrated solution, successful lab scale experiments allowed us to define the basic guidelines for both the recovery of Ag from the anolyte and the online separation and regeneration of silver halides that could be formed when halogenated organic materials are being oxidised. The basic principle relies on the reaction between silver halides and H_2O_2 at high pH and controlled temperature and concentrations. We also carried out laboratory experiments to assess the oxidation of bitumen by electrochemical mediation. Although reactions take place, they appear to be quite slow. Enhancing the contact between reactants is a key point. This requires coupling with complementary techniques (e.g. extraction, pyrolysis).



Schematic design of the sodium treatment process

The selective extraction of cobalt from radioactive effluents

SCK•CEN decided on the basis of new information on the characteristics of the MEDOC effluents and a shortage of manpower, not to continue the studies on the selective extraction of Co out of the effluents. Nevertheless, significant results were achieved during this study. We developed a new synthesis route for corroles, obtaining yields three times higher than an alternate route that has been recently patented in Israel. The first three steps of the synthesis are carried out advantageously in one single vessel, leading to dipyrromethane 1,9-dicarboxilic acid. Next, corrole is being formed through adequate ring closure carried out in the presence of triphenylphosphine and cobalt (II) acetate. Due to the structural similitude of corrole and vitamin B12, we expect the ligand to be particularly efficient for extracting traces of Co from effluents containing large amounts of other metals (Fe, Ni and Cr). We won the SCK•CEN award 2000 for the non-university final year thesis with this work (see Annexes). These studies caused the discovery of an unknown crystallographic form of triphenylphosphine oxide, which is a remarkable sideline result. In this framework, we also prepared the future study of a membrane in which our selective molecule could be embedded through examining the behaviour of a polysulfone membrane containing dead cell membranes of *Enterococcus hirae* (Formerly called *Streptococcus faecalis* or *faecium, E. hirae* is a facultative anaeorobe. It has a simple metabolism and can generate ATP only by glycolysis. This bacterium has therefore been used for bioenergetic studies for over 30 years. Electrochemical impedance spectroscopy (EIS) measurements are affected by a poor reproducibility, unless particular precautions are used (e.g. efficient preliminary outgassing, characterisation in the presence of one single heavy metal in the solution (Ni^{2+}), careful examination of the trend in function of time). Cyclic voltammetry indicates that graphite as well as vitreous carbon may be used as electrode material.

Perspectives

The activities at BR3 will continue with the decommissioning of the main components (steam generator, pressuriser, pump housings) of the primary circuits using the high water jet cutting technique. The primary loop piping under the operating deck will be, after dismantling, decontaminated in the MEDOC facility. The MEDOC facility could be adapted to allow the in situ decontamination of the steam generator and the pressuriser. This scenario will be submitted to the technical liability manager early in 2001.

Concerning the waste treatment processes we are developing, ours goals are to treat the 180 l of tritiated methanol and to demonstrate the validity of our process for the treatment of alkali metals by the complete oxidation of first 1 and, next 10 kg of non-radioactive sodium. Further, we aim at completing the development and the validation of the models describing the oxidation of alkali metals and the oxidation of organic waste through Ag^{2+}. We further expect to address clearly the technical questions raised by bitumen as waste form and to improve the completeness of the oxidation of organic waste through Ag^{2+} process and the MEDOC process by minimisation and treatment of the secondary waste.

We will more and more develop our role as consultant and/or industrial partner in decommissioning and waste minimisation activities.

Partners

-	Belgatom (Belgium)
-	FRAMATOME (France)
KULeuven	Katholieke Universiteit Leuven (Leuven, Belgium)
OPT	Ontario Power Technologies (Canada)
RUCA	Rijksuniversitair Centrum Antwerpen (Antwerp, Belgium)
UIA	Universitaire Instelling Antwerpen (Antwerp, Belgium)
ULg	Service d'Electricité Appliquée (Prof. W. Legros), Université de Liège (Liège, Belgium)
VITO	Vlaamse Instelling voor Technologisch Onderzoek (Mol, Belgium)
WTCB/CSTC	Wetenschappelijk en Technisch Centrum voor het Bouwbedrijf (Belgium)

Sponsors

IRI	Institute of Research and Innovation (Japan)

Customers

ABB-ATOM	Nuclear department of ABB (Sweden)
BN	BELGONUCLEAIRE (Dessel, Belgium)
CRIEPI	Central Research Institute of Electric Power Industry (Japan)
EC	European Commission, DG Environment (Brussels, Belgium)
JGC	Japan Gas Company (Japan)
MEZ	Ministry for economic affairs (Brussels, Belgium)
NECSA	Nuclear energy Corporated South Africa
ONDRAF/NIRAS	Belgian Agency for Radioactive Waste and Enriched Fissile Materials (Brussels, Belgium)
RAO-Agency	Radwaste Agency (Slovenia)
RUG	Universiteit Gent (Belgium)

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