FOREIGN TRAVEL REPORT: VISITS TO UK, BELGIUM, GERMANY, AND FRANCE TO BENCHMARK EUROPEAN SPENT FUEL AND WASTE MANAGEMENT TECHNOLOGY

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EXECUTIVE SUMMARY

The ICPP WINCO Spent Fuel and Waste Management Development Program recently was funded by DOE-EM to develop new technologies for immobilizing ICPP spent fuels, sodium-bearing liquid waste, and calcine to a form suitable for disposal. European organizations are heavily involved, in some cases on an industrial scale in areas of waste management, including spent fuel disposal and HLW vitrification. The purpose of this trip was to acquire first-hand European efforts in handling of spent reactor fuel and nuclear waste management, including their processing and technical capabilities as well as their future planning. Even though some differences exist in European and U.S. DOE waste compositions and regulations, many aspects of the European technologies may be applicable to the U.S. efforts, and several areas offer potential for technical collaboration.

Vitrification Technology

High-level waste resulting from reprocessing is being processed in an electrical induction-heated melter by the French and British using the Atelier de Vitrification La Hague (AVH) technology developed by the French for the La Hague Reprocessing Plant. The longest operation has occurred at La Hague in the R7 Vitrification Plant where 1350 canisters (150-l volume) of glass have been produced in 3 years. The induction heating is applied to the metal melter which in turn heats and melts the glass inside. Temperatures up to 1130°C must be withstood by the metal melter body, resulting in relatively short melter lifetimes. The French are experiencing average melter lifetimes of approximately 1650 hr, are projecting a melter life of 2000 hr beginning in 1993 and average 2 days per replacement of a failed melter. Their goal is a 5000 hr average melter life in five years. The British have spent £ 10 Million ($15.5 Million), which includes thousands of hours of testing, in implementing the AVH technology with French assistance. Cost of Vitrification Facilities was £ 240 Million ($375 Million). Since 1991 they have experienced four melter failures due to corrosion, a failed melter nozzle and a rotary calciner bearing failure. They are experiencing an average melter life of 1400 to 1450 hr versus a design basis of 2000 to 3000 hr, and a failed melter
replacement time between 6 days to 6 months. The lengths of melter replacement time they feel are directly related to the availability and reliability of auxiliary equipment such as cranes and manipulators, and they are striving for a two to three week changeout period. The shorter changeout time in the French facility was attributed by the French to personnel training and their organizational structure and management. A longer melter life is believed by the French to be directly related to temperature control, careful control of melter material molecular grain structure, use of an inert argon atmosphere above the glass melt and optimized temperature distribution within the melter. The French feel their increased melter life goals are achievable primarily by improved temperature distribution within the melter through use of heating fins. The AVH technology is limited by heat transfer, vessel strength and corrosion resistance. The progress of AVH operation at Sellafield and La Hague should continue to be monitored for applicability to ICPP wastes.

The importance of up front operational considerations in a facility design was vividly illustrated in comparing the French and British designs for transfer of vitrified glass canisters from the loading/decontamination cell area to the interim storage wells. In the French facility, this was accomplished by a moveable cask on the same elevation, while in Sellafield, the glass canister was loaded in a cask, moved to a higher elevation for unloading into the storage tubes.

The Germans/Belgians have successfully vitrified liquid high-level waste generated from past operation of the Eurochemique Reprocessing Plant using Joule-heated Liquid-Fed Ceramic Melter (LFCM) technology as a radioactive pilot demonstration. The equivalent of 1300 150-l canisters was produced over six years of operation at approximately one-half of the capacity of the industrial-scale R7 La Hague Vitrification Plant. Melter life for this effort averaged three years and changeout time involved ten weeks. Principal drawbacks with the melter designs employed to date is the ability to handle noble metals and the need for controls on levels of metal constituents. The noble metals can concentrate near the melter bottom leading to increased localized current densities and attendant electrode corrosion. High levels of
metal constituents can increase viscosity and reduce molten glass flow, stimulating the need for increased heating and the potential for electrode and pour spout damage. The Germans are developing and testing at KfK an alternate Joule heated melter that employs a steep sloped lower region and bottom drain with relocated electrodes. They are also testing Joule heated melter designs for China and Savannah River/Hanford programs. These efforts should be closely followed for potential application in treating ICPP waste.

The French currently are developing a cold crucible melter concept that was originally patented in 1925. This concept uses electric induction heating directly to the glass inside the melter. The induction energy passes through openings in the axial segmented cylindrical metal melter body in which each segment is cooled by water flow. In order that the induced current can heat the glass, some glass must be melted initially by other means, such as plasma torch or microwave. Since the metal melter walls are cooled, a layer of solid glass forms on the inside surface of the melter, protecting the melter walls from the corrosive molten glass and offgases. Temperatures up to 1500°C can be achieved, as opposed to 1130°C for the AVH melters. Glass can be continually poured through a bottom drain. This melter design has also been used for melting stainless steel and zircaloy fuel cladding hulls. As with the glass, the induction current is directly applied to the hulls through the gaps in the cooled metal melter segments. As new hulls are added, the melt can be extruded using a moveable piston at the bottom, forming a metal ingot. Small amounts of fluxing salts are required in the process and produce a slag, separating some of the radioactive elements. The process offers several interesting possibilities for metal recycle and vitrification, and this on-going French effort should be closely followed.

Alternative Approaches to Dispositioning of Spent Fuel

The visits identified that the British and French have committed and are planning to continue committing significant capital resources to reprocessing of spent nuclear fuel, did not discuss any efforts to evaluate different alternatives and feel that the current economics of reprocessing and plutonium recycle are favorable to a direct disposal option. Britain is currently
receiving considerable public pressure to build a new £ 50 Million Krypton Removal Plant prior to commencing operation of a new Thermal Oxide Reprocessing Plant planned for late 1992 early 1993. Germany on the other hand is proceeding with developing to maturity a spent fuel direct disposal option for use when reprocessing is not available or economically unjustified. The German studies of various disposal options conclude that the cost difference between options is mainly caused by changing types and numbers of containers accompanying changes in emplacement concepts. Continued follow-up of the British, French and German effort is appropriate for information useable in the U.S. programs.

Other Waste Management

Other waste management topics which were applicable to the ICPP program and should continue to be monitored include the following:

There is a strong effort at KfK by the Germans on repository performance assessment and on spent fuel and waste form testing. Metal melting/decontamination technology appears to have been successfully applied by the British, French, and Germans. In these countries there are de minimus values of residual radioactivity which allows for useful recycle of much material. Decontamination and decommissioning technologies are also being actively implemented by all countries. The Germans are pursuing drying of Intermediate Level Liquid Waste (ILLW) and have achieved through drying of a ILLW concentrate a salt with a five fold volume reduction compared to the concentrated ILLW and a ten fold volume reduction compared to cemented ILLW product. While this technique might be useful in principle for treating ICPP sodium waste, a large increase in throughput would be required for it to be practical. Cement/grout technology is being actively used at all sites. Radionuclide separations technology is being developed at France, Germany and UK. The TRU waste incineration tests are ongoing at Marcoule, France.
Follow-On Actions

- BNFL will forward available information on glass quality as a function of residence time in the melter.

- BNFL will furnish any available non proprietary information on specification of waste characteristics, vitrified waste glass characteristics, vitrification process additives, and wet scrubbing efficiency for NOx absorption.

- BNFL will furnish available information on influence of grouting on material solubility when exposed to liquid after grouting.

- BNFL to forward a package of information portraying their remote capabilities.

- WINCO to contact Malcolm Saunders of the UK concerning British decontamination development efforts.

- BNFL to forward a package of information portraying their Decontamination/Decommissioning capabilities.

- WINCO to continue to follow German effort on direct disposal of spent fuel.

- WINCO to continue to follow French and German Vitrification melter development efforts.

- WINCO to follow French efforts on cold crucible melter concept for applicability.

- WINCO to provide French with available information in pyrochemical processing.
• WINCO to investigate potential modifications of the German Intermediate Level Liquid Waste Drying Technology for use in processing sodium waste.

• SGN to forward available waste acceptance specifications for glass, cement, and bitumen
INTRODUCTION AND PURPOSE

The ICPP WINCO Spent Fuel and Waste Management Development Program recently was funded by DOE-EM to develop new technologies for immobilizing ICPP spent fuels, sodium-bearing liquid waste, and calcine to a form suitable for disposal. The program also initiated an effort in developing decontamination techniques which minimize sodium waste generation and technologies for decontamination and recycling contaminated metals. To help with effective identification and implementation of the applicable technologies, a benchmarking effort of U.S. and foreign programs is needed.

European organizations are heavily involved, in some cases on an industrial scale in areas of waste management, including spent fuel disposal and HLW vitrification. French/UK vitrification, including other associated waste processing for intermediate level and LLW immobilization/disposal, is operating on an industrial scale. German vitrification technology has been demonstrated on Eurochemique HLW at Dessel, Belgium. The Germans also have an extensive development effort in spent fuel disposal using a long-life cask and repository performance assessment. German/French have ongoing R&D in waste form characterization and tests on repository behavior. Concerns have been expressed by U.S. Senator Grassley in reviews of the DOE vitrification program that foreign technology has previously not been adequately considered by DOE, and a request has been made that DOE adopt applicable technologies where possible.

The purpose of this trip was to acquire a first-hand knowledge of pertinent European efforts in technology and application related to DOE spent fuel and nuclear waste management, as well as an understanding of their capabilities which may assist in closure of the United States nuclear fuel cycle in a cost-effective manner. The itinerary included the Sellafield site in the UK, the PAMELA vitrification facility in Belgium, the Karlsruhe Nuclear
Research Center (KfK) in Germany and the La Hague and Marcoule sites and SGN headquarters in France. During each visit an overview of capabilities was obtained and detailed discussions with key technical personnel conducted.

The trip provided insight into European spent fuel and waste management technology and capabilities. This report presents our observations of selected European technologies, but at this time, applicability to ICPP or the defense waste streams can only be estimated. For example, most of the European high-level waste contains a much higher fission product and actinide content (up to 50 wt %), while ICPP HLW calcine contains high levels of nonradioactive materials (99 wt %) and lower fission product/actinide content (less than 1 wt %). These differences may require process adjustments and testing before implementation. Differences in regulatory release limits also may affect technology applicability, e.g. all countries which were visited allowed a "de minimus" level of contaminated metals to be recycled to the commercial market (not allowed in the U.S.).

The following section provides detailed descriptive information associated with each visit. This is followed by a section listing personnel contacted at each site and a section giving follow on action. A summary of key observations, conclusions, and follow-on action associated with the trip is provided in the Executive Summary.
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ITINERARY

Tuesday October 27 Sellafield, UK
Wednesday October 28 BNFL, Sellafield Site, UK
Thursday October 29 BNFL, Sellafield Site, UK
Friday October 30 Belgoprocess, Dessel, Belgium
Saturday October 31 Karlsruhe, Germany
Sunday November 1 Karlsruhe, Germany
Monday November 2 KfK (Nuclear Research Center), Karlsruhe, Germany
Tuesday November 3 Cherbourg, France
Wednesday November 4 COGEMA, La Hague, France
Thursday November 5 CEA, Marcoule, France
Friday November 6 SGN, Paris, France

TRIP COSTS

Total Cost of trip was approximately $ 8,000
The visit to the British Nuclear Fuels, plc (BNFL) in Sellafield provided information on the background and capability of the facility process flow, its classification and handling of waste types, and focused discussions on key items such as waste treatment, robotics and decontamination.

**Background and Capability**

Sellafield, a 700 acre site, is located in the northwest corner of England along the coast of the Irish Sea south of Whitehaven and is operated by BNFL. The BNFL in its present form was created in 1971 out of the former Production Group of the United Kingdom Atomic Energy Authority with a head office and engineering design center at Risley (2600 employees), fuel manufacturing facilities at Springfield (3250 employees), uranium enrichment facilities at Capenhurst (1300 employees), a nuclear power station at Chapelcross (650 employees), and the Calder Hall nuclear power station and fuel handling, reprocessing, and waste management facilities at Sellafield (7,900 employees). BNFL became a public limited company in 1984 and achieved an operating profit of £ 339 Million ($530 Million) on a turnover of £ 1,042 Million ($1,625 Million) during the 1990/91 financial year.

The Sellafield Calder Hall nuclear power station consists of four 60 MW Magnox type reactors which like other first generation British reactors utilize as a reactor core natural uranium metal cast into rods and fitted into sleeves made of a magnesium alloy. More recently installed reactors in Britain include Advanced Gas Cooled Reactors (AGRs) and Pressurized Water Reactors (PWRs) that utilize pellets of enriched uranium which are loaded into stainless steel or zirconium sleeves.

The initial Sellafield reprocessing facility that began operation in 1952 was designed specifically for the reprocessing of Magnox fuel elements. Sellafield’s current Magnox fuel reprocessing plant started operation in 1964.
and has undergone a comprehensive program of refurbishment and improvement which, together with continuing upgrade efforts, is anticipated to allow operations into the 21st Century.

A new £300 Million Fuel Handling Plant for receipt, storage and preparation of Magnox fuel for reprocessing was opened in 1985. The fuel storage water basin consists of three bays each with a capability of storing 1,680 tonnes of Magnox fuel. The pools are concrete covered with an epoxy coating. Fuel is stored in closed containers to prevent any contamination of the pool surfaces and water. Also commissioned in 1985 was a £200 Million Site Ion Exchange Effluent Plant (SIXEP) which filters water from the original outside fuel storage basins and associated facilities, using a Zeolite (clinoptilolite) ion exchange material to remove cesium and strontium prior to discharge to the Irish Sea.

To accommodate a current £9,000 Million backlog of orders from customers in nine different countries, construction is nearing completion on a £2.8 Billion (£1,850 Million base facility cost and £950 Million supporting facilities) Thermal Oxide Reprocessing Plant (THORP) which will reprocess fuel from PWRs and AGRs. The THORP facility has a design throughput capacity of 7,000 tonnes for the first ten years versus a current booked backlog of 6,000 tonnes for the first ten years. THORP is nearing completion and is expected to be commissioned in late CY92 early CY93. Currently BNFL is receiving considerable public pressure to build a £50 Million krypton removal plant prior to increasing planned discharges associated with operation of THORP. Building such a plant would have a significant adverse affect on THORP projected economics.

A new £240 Million Vitrification Plant was officially opened in 1991 which uses French technology of rotary kiln calcination followed by melting in an electrical induction heated metal melter to convert concentrated high activity reprocessing raffinate liquid waste into a stable glass form with a volume reduction of two thirds (see Figure 1). The stainless steel containers of vitrified waste glass are placed in an air-cooled storage facility adjacent
High-level liquid waste, currently in tank storage, is transferred via a pipebridge into the Vitrification Plant's highly active liquor tank. It then goes through the vitrification process which consists of the following stages:

1. The liquid is evaporated and dried into a powder.
2. The dried powder is fed into a melting pot together with glass making material. 25% powder (waste) to 75% glass. The powder and glass fuse together.
3. The molten mixture is poured into a container.
4. The containers are transferred to an air-cooled store and stacked 20 high in stainless steel tanks.

Figure 1. Schematic Layout of Sellafield Vitrification Plant
to the Vitrification Plant where they will be stored for at least 50 years to allow heat output and radioactivity to decay, prior to placement in a permanent British repository.

BNFL has underway an additional program requiring an investment of £600 Million for three plants to encapsulate Intermediate Level Waste (ILW) (spent fuel metal cladding sections, certain contaminated equipment, and sludges from reprocessing/clean up activities) into a concrete matrix for storage at Sellafield site awaiting final deep geologic disposal. The first plant was opened in 1991 and the others are scheduled for operation during 1992.

Process Flow

The process flow at Sellafield begins with receipt and water storage of Magnox, AGR and PWR fuels in 50 tonne cuboid Magnox casks and 110 tonne cylindrical casks for PWR fuel. To minimize storage volume, the 36 AGR fuel elements are consolidated by removal from their outer graphite sleeve and attendant stainless steel (SS) support structure and combining with 72 other AGR fuel elements for storage in a cylindrical container prior to reprocessing. The storage container for the consolidated elements has the same configuration and volume as the original graphite sleeve.

The graphite sleeve with approximately 200 mSv activity and cobalt contamination is crushed using a two-stage process. During the first stage the graphite sleeve is normally broken into three - 120° segments allowing removal of the SS support structure. In the second stage, these segments are broken into roughly 6" long segments of various circumferential lengths. The crushed graphite segments are loaded into lacquor-coated carbon steel vessels awaiting repackaging once disposal/encapsulation requirements are established. Effort is under-way to change the lacquor-coated carbon steel vessels to a comparable SS vessel. The SS support structure is chopped into pieces for encapsulation and disposal as an ILW.
It was noted that the graphite sleeve experiences some shrinkage and distortion which is a function of its location in the reactor core and fuel burn up. Also some difficulty is experienced in removal of the fuel pins due to bowing and locking of the fuel elements in the support grid, which was considered to be the most pronounced affect. Occasionally the graphite sleeves crack and cause difficulty in handling.

After a minimum storage period in the water basins which is maintained at 15° C, the Magnox fuel magnesium alloy sleeve is stripped from the cast uranium metal core, and the core is transferred to the reprocessing plant for dissolution and separation of reusable uranium and plutonium. After a minimum storage time, AGR and PWR elements will be reprocessed in the new THORP facility. The reprocessing flowsheet begins with chopping the cylindrical fuel elements into roughly 2" lengths or rounds which then flow to a dissolver where the core fuel is dissolved out of the sleeve, followed by separation of reusable uranium and plutonium. The shavings from the Magnox fuel and the AGR and PWR sleeve rounds are sent to facilities for encapsulation in concrete as ILW. Raffinate from the dissolution is concentrated and processed as high-level waste (HWL) in the Vitrification Plant.

**Waste Types**

The British classify nuclear waste in one of three categories.

**High Level Waste (HLW)**

HLW is the acid solution raffinate resulting from fuel reprocessing which contains mainly fission products and actinides other than plutonium. British reprocessing has generated about 1,300 cubic meters of HLW which is scheduled for vitrification and ultimate disposal in a permanent repository.
Intermediate Level Waste (ILW)

ILW has an activity greater than a LLW cutoff and is not suitable for shallow land disposal. It consists of spent fuel sleeve sections, certain contaminated equipment, and sludges from reprocessing/clean-up activities. The waste is being or is planned to be encapsulated in a cement inside a SS drum and stored in interim facilities pending siting and construction of a long term geologic repository.

Low Level Waste (LLW)

LLW comprises effluents and solids which are only slightly radioactive and suitable for near-surface disposal. Solid LLW consists of items such as towels, plastic sheeting, cardboard and effluents of liquid and gaseous discharges to the environment. A 300-acre land fill disposal site at Driggs, just south of Sellafield, constitutes the existing LLW repository, and effluent emissions are controlled consistent with regulatory requirements.

Focused Discussions

The focused discussions centered on alternative approaches to dispositioning of spent fuel, waste treatment, robotics, and decon.

Alternative Approaches to Dispositioning of Spent Fuel

BNFL has a large investment of £2,800 Million in THORP and supporting facilities to reprocess spent nuclear fuel, with sufficient current orders from utilities for 10 years of operation. Future orders will likely depend on the economics of continuing to reprocess versus changing to disposal of fuel. An inquiry was made concerning any BNFL evaluations that have been made on current economics associated with reprocessing versus direct disposal of spent fuel. Although no evaluations were presented, they took the position that reprocessing is the appropriate responsible approach irrespective of the value
of recovered product and associated economics. It will be interesting to see how efforts in other countries toward direct disposal influence their future backlog of reprocessing orders.

Waste Treatment - Vitrification

After the French constructed and successfully demonstrated their vitrification process at AVM at Marcoule and AVH at La Hague, the British decided to abandon their vitrification development efforts and adopt the French technology which was projected to allow for production scale vitrification operations at Sellafield several years earlier than originally forecast. A contract was signed between BNFL and Société Générale pour les Techniques Nouvelles (SGN) to transfer the French Atelier de Vitrification de La Hague (AVH) technology and to involve SGN in the design of the Sellafield plant. A series of trials took place at Marcoule where the process proved to operate successfully with simulant of British HLW. A similar full-scale test facility was constructed and operated at Sellafield. Although the approach was based on a tried and proven French Technology, BNFL spent on the order of £10 Million developing the process for British application with the principal focus on Magnox waste streams. It is anticipated that THORP waste streams will be bracketed by the Magnox streams. BNFL identified that the glass formation was relatively tolerant of chemical composition and that they had not completely investigated their currently projected range of burn up and the attendant potential influence of additional noble metals, etc. There is a 15-wt% level of fission product oxides in their glass as part of the waste oxide content in the glass of 25 wt%.

BNFL noted that there are three major areas where their plant design differs from that of the French - cell layout, pretreatment/liquid handling, and storage.
Area 1 - Cell Layout

Changes were made to the cell layouts to improve maintainability, alter or incorporate redundant components for increased lifetime and reliability, and modify waste streams fed to the process. BNFL noted that while they had made significant modifications, they wish that they had made more especially with respect to reliability and availability of auxiliary equipment used to maintain equipment and operate the facility, such as cranes and manipulators. This item was felt by BNFL to be the key item associated with plant productivity.

Area 2 - Pretreatment/Liquid Handling

Changes were made to the process to reduce maintenance by eliminating moving parts through use of flow movers, fluid agitation and duplication of components that could not be built for the life of the plant. BNFL felt that further improvements could have been made to the process off-gas system through the use of Reverse Flow Diverter pumps, which is a pumping concept with no moving parts.

Area 3 - Storage

The Sellafield HLW canister (SS containers of vitrified waste glass) storage facility stores the HLW canister ten high in a cylindrical tube that is surrounded by an outer tube which allows for natural convection cooling via a 3-4" radial gap versus forced convection cooling used by the French. The storage facility is designed to accommodate a 2.5 kW heat loading per container, and it is planned to store the canisters a minimum of 50 years in the facility prior to movement to a permanent British repository. It was also identified that the 2.5 kW heat loading maintains the glass with a center line temperature less than 500° C. The existing storage facility will accommodate a total of 80,000 containers. BNFL identified that in retrospect they should have adjusted the elevations between the canister filling/decon area and canister storage area. This would have reduced handling complexities and simplified canister transporter requirements.
The Sellafield Vitrification Plant has two AVH-sized lines, each with a 25 kg/hr capacity. In each line HLW is continuously fed into a 3% sloped rotating SS tube rotary kiln calciner with zoned resistant heating and a rabble bar to break up any large deposits. Calciner inlet temperature is 650°C and outlet temperature is 850°C with an average temperature of 725°C. The calciner feed is conditioned with lithium to improve glass loading of cesium and strontium, and sugar solution is added to retain ruthenium.

Calcine from the calciner and glass frit are continuously fed in a one-to-three ratio into an elliptically cross sectioned Inconel 601 induction heated melter, whose outside skin temperature is maintained at approximately 1150°C. An air sparge is used for stirring, with its primary function to induce uniform heating rather than constituent mixing. The molten glass liquid inside the melter has a crusty layer of entering calcine and glass frit which also provides for heat shielding of the upper melter. A roughly four-hour residence time is experienced by the entering calcine and glass frit. BNFL indicated that information on glass quality as a function of resident time in the melter was available and that they would forward this information to WINCO. Every eight hours 1/2 of a HLW canister volume is removed from the melter over a weir which is designed to maintain a 70-kg heel. Discharge to the canister is controlled by melting and reformation of a glass plug in the discharge line, using a separate induction heater. The exit temperature of the melter product is 1050°C as it enters the HLW canister which is heated to 650°C. The product container is approximately 42 cm in diameter and 1.3 meters high and holds about 400 kg of glass (150 liters).

Following receipt of two melter discharges, the canister is allowed to cool naturally. A fair amount of cracking occurs during the cooling process, which increases the glass surface area by a factor of 10-20. Impeded cooling could increase devitrification and affect leachability. Following cooling, the container is fitted with a lid and sealed by an automatic fusion welding technique. It is then decontaminated and sent to interim storage as previously described.
BNFL has experienced to date four melter failures due to corrosion and one pouring nozzle failure due to overheating. The average life time between melter failures due to corrosion has been conservatively 1400 hrs versus a projected 2,000-3,000 hour operating design life. Each corrosion failure has started with a pin hole type failure identified by a loss in vacuum which is followed by, in roughly a 12-24 hours time period, a cracking opening of the melter. Times to replace the melter have varied between as little as 6 days to as much as 6 months. BNFL expects to get melter replacement into a consistent 2-3 week time period and emphasized the importance of reliability and availability of auxiliary equipment such as cranes and manipulators. It was also identified that a water-cooled rotary calciner bearing had failed since operation began in 1991.

BNFL is currently pursuing use of an improved melter material that provides the needed levels of conductivity and corrosion resistance. A modified 690 alloy is being pursued. Difficulties in translating results from the prototype melter to the process melter were expressed, and it was noted that heat transfer, vessel strength and corrosion resistance are limiting to the AVH technology. BNFL is not pursuing microwave heating as used by the Japanese.

The Vitrification Plant off-gas treatment system consists of a primary treatment process, one for each process line, followed by treatment in a secondary off-gas system. As the primary treatment, off-gas from the calciner is processed at 100 m³/hr through a liquid mist dust scrubber followed by a condenser and by a wet scrubber for absorbing NOₓ. In the secondary system the effluent passes through two parallel electrostatic precipitators, a chilled water bubble column or wet scrubber, and HEPA filtration prior to discharge.

Discussions/questions of BNFL concerning specific information pertaining to specifications on waste characteristics, vitrified waste glass characteristics, process additives, effects of melter residence time on glass
quality, and wet scrubbing efficiency for NO\textsubscript{x} absorption resulted in an agreement to furnish available non proprietary information in the near future.

**Waste Treatment - Encapsulation**

The shavings/pieces from the stripping of the Magnox fuel magnesium alloy sleeves are placed in a container with two screen-like tubes. Water is withdrawn from the container using the one tube and the remaining tube is used to measure liquid level. An anti-floatation device is installed in the top of the container, and the container is vibrated during pouring and initial curing of a liquid cement mixture to achieve a relative even distribution of the magnesium alloy shavings/pieces within the cement. Following a curing period, a final concrete layer is added, a lid is welded to the container and the container is scrubbed to remove any contamination prior to being sent to interim storage awaiting disposition in a final repository. BNFL identified that process development required significant investigation to achieve an acceptable distribution of the shavings/pieces within the final cured concrete as well as careful considerations to accommodate the pyrophoric nature of the magnesium alloy shavings. It was also identified by BNFL that information was available illustrating that grouting decreases the solubility of material that is subsequently exposed to liquid, and they agreed to forward this information to WINCO.

**Robotics**

BNFL has considerable experience in robotics and remote handling, including in-cell mechanical process and production equipment. Their basic approach is to utilize where possible off-the-shelf robotic components and to extend the capabilities of such components or develop new capability only as the need arises to meet specific needs. Two specific new systems currently under development were toured and discussed during the visit as follows:
REP (Repair) MAN

REPMAN is a robotic arm that is being developed to install sleeves in selected pipe lines that are connected to the North Reprocessing Dissolver vessel for the Magnox reprocessing facility. Since 1986 BNFL has been monitoring robotically selected welds in pipe lines that branch from the dissolver vessel. To extend the life of the dissolver system it was decided to install, using a TIG welding process, sleeves that would cover certain welds in 3/4" to 3" diameter lines connecting to the vessel and to plug other lines no longer needed for operation. REPMAN, a five degree of freedom (3 hydraulic, 2 electrical) robotic arm with a 15 kg design weight capacity, is being developed for insertion into the vessel through a 6" diameter line to accomplish the required repairs. Actual development began in 1987 and, after having been put on hold for a period of time, has progressed to performance of the desired repairs in a mockup with a total expenditure to date of over £ 4 Million. The REPMAN software utilizes operator location control and a series of pre-programmed routines for performance of specific activities such as welding of a sleeve.

Raffinate Pipe Adjustment (Raff Man)

Raff Man is a remote process being developed to remotely reroute the raffinate line from the Magnox South dissolver vessel to the North reprocessing separation columns. To accomplish this it will be necessary to work remotely through an unshielded 2' diameter cell penetration to remove a section of pipe from an existing line, cap one of the resulting pipe ends, and weld new piping to the other, connecting it to a new line penetrating the cell. Work began on this effort in the late 1988 to early 1989 time frame and has progressed to the mock-up demonstration phase at a cost to date of over £ 4 Million. Remote modification was selected as the best approach rather than decon and direct modification. BNFL identified that they would forward a package of information concerning their complete robotic capabilities.
Discussions concerning decontamination technologies centered on BNFL's approach to decontamination and project management of specific decontamination efforts. Personnel knowledgeable of British development efforts were not available, and it was suggested that Malcolm Saunders (011-44305-251888 ex. 2135) of the Atomic Energy Authority be contacted concerning development efforts. Currently BNFL is decommissioning their original weapons grade material production facilities located at Sellafield. Decontamination of one of the facilities has necessitated use of robotics due to a production reactor fire that occurred in 1957 which spread contamination. Their general approach to decontamination is to begin decontamination as soon as possible for plutonium contamination, in order to minimize high gamma energy hazards from the americium daughter decay product and to use hands-on disassembly only after a reduction in radiation fields to acceptable levels. As in the case of robotics, available technology and proven approaches are utilized to reduce contamination to the point that the waste may be disposed of as LLW.

BNFL identified that they would pull together a package on their D & D expertise and capabilities which could serve as a basis for any desired follow on discussions.
The Belgoprocess Facility is located at Mol, Belgium and contains radioactive waste handling, processing, and storage facilities. The site is operated by Belgoprocess which is a private-owned subsidiary of Niras/Ondraf, a Belgium state agency. During the 1985-to-1991 time frame, the PAMELA vitrification facility was operated at the site to process approximately 907 m$^3$ of liquid waste into a glass product using a Joule-heated ceramic melter. Two melters were required at an average life of three years each. During operation of PAMELA, the site employed a staff of 300 people with an annual 1.6 Billion BF ($50 Million) operating budget. Currently, the first melter is stored in an adjacent cell awaiting disassembly, and the second vitrification melter is being disassembled and packaged for disposal. Of principal interests in this visit were the PAMELA design features and operating experience.

PAMELA Melter Design

The basic flowsheet for the PAMELA Vitrification Plant is given in Figure 2. The plant utilizes a continuous liquid-fed ceramic melter that uses 8 electrodes (4 pairs of 2 electrodes) with a total design heating capacity of 75 kW and installed electric power of 130kVA to Joule heat the glass to immobilize a liquid waste/glass frit mixture. A cut-away view of the melter is shown in Figure 3, and selected melter construction data is provided in Table 1. The melter brick is a high chromium ceramic required to withstand the highly corrosive molten glass. The melter has a 20-liter/hr feed rate, a 300-liter capacity and generates approximately 400 kg/day of glass product, sufficient to fill one 150 l glass waste container the same size as used by the French and English. This production rate is comparable to one of the AVH process streams—at 25 kg/hr. Glass product is removed using either a bottom gravity feed nozzle with a design throughput of 75-80 kg/hr, or an air lift feed overflow nozzle with a design throughput of 100 kg/hr. Some agitation of the melter contents is provided by bubbling nitrogen.
Figure 3. Cut-Away Schematic of PAMELA Liquid-Fed Ceramic Melter.
Table 1: Construction data of the K4-Ceramic PAMELA Melter

<table>
<thead>
<tr>
<th>Description</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>OUTER DIMENSIONS</td>
<td>2 X 2 X 2m</td>
</tr>
<tr>
<td>OVERFLOW</td>
<td>0.7 X 0.9 X 0.8m</td>
</tr>
<tr>
<td>TOTAL WEIGHT (incl. glass)</td>
<td>18.8 tonne</td>
</tr>
<tr>
<td>REFRactories MATERIAL</td>
<td>1.4 m$^3$; 5.2 tonne</td>
</tr>
<tr>
<td>INSULATION MATERIAL</td>
<td>5.4 m$^3$; 7.3 tonne</td>
</tr>
<tr>
<td>METALLIC BUILT-IN DEVICES</td>
<td>2.0 tonne</td>
</tr>
<tr>
<td>STAINLESS STEEL CONTAINMENT</td>
<td>3.5 tonne</td>
</tr>
<tr>
<td>MELTER BATH SURFACE</td>
<td>0.77m$^2$</td>
</tr>
<tr>
<td>MELTER BATH CONTENT</td>
<td>0.8 tonne glass</td>
</tr>
<tr>
<td>INSTALLED ELECTRIC POWER</td>
<td>130 kVA</td>
</tr>
</tbody>
</table>
Melter Operation

Initial hot testing of the PAMELA Plant began in late June 1985, and the beginning of processing occurred in October 1985. During the period October 1985 through July 1991, the plant processed approximately 47 m$^3$ of Low Enriched Waste Concentrate (LEWC) generated from reprocessing of Power Reactor Fuel and approximately 860 m$^3$ of High Enriched Waste Concentrate (HEWC) generated from reprocessing of aluminum-clad Research Reactor Fuels. After approximately one year of operation, the bottom 15 mm diameter nozzle plugged. This plugging was felt by the PAMELA operations personnel to possibly be associated with the high concentration of noble metals in the power reactor fuel. The total 47 m$^3$ of LEWC contained approximately 90 kg of noble metals. The noble metal fission products, mainly ruthenium (Ru), rhodium (Rh), and palladium (Pd), have a low solubility in glass and precipitate as oxides or metal alloys in the melt. Due to their high density, they settle and concentrate on the bottom as a more viscous liquid with higher electrical conductivity than the rest of the glass. This can lead to a local increase in electrode current density through the noble metal phase with a corresponding decrease in current density through the rest of the melt, requiring increased operating voltage to maintain the glass temperature. The higher localized current density causes electrode corrosion. The PAMELA operations personnel noted that noble metals are not tied up chemically within the melter and stated their belief that the nitrogen agitation is not sufficient to keep the denser noble metal oxides and alloys from settling. Figure 4, which identifies a sharp rise in the voltage/current ratio during processing of the high noble metal LEWC Power Reactor Fuel, was provided to substantiate their belief concerning high noble metal influence on operating characteristics. Rather than change out the melter, it was decided to continue operation using the nitrogen air lift and the overflow nozzle for canister pouring. This continued until the April-May 1990 time period when the plate boundary separating the melter's main waste liquid/glass frit solution from the overflow channel solution failed. Since the quality of the solution being withdrawn from the melter could no longer be assured, it was decided to suspend operations and replace the melter.
Figure 4. Voltage/Current Ratio and Accumulated Noble Metal Inventory in the Operation of the PAMELA Melter.
Replacement of the initial melter with a second melter took 10 weeks following which operations were resumed. Early on during operation of the second melter the bottom nozzle again plugged. The PAMELA operations personnel did not believe this plugging to be associated with noble metals since their levels are fairly low in HEWC and did not offer any other basis. In subsequent discussions at Karlsruhe, a possible explanation was presented related to the high aluminum content of the melt (see the following section). Discharge was switched to the overflow nozzle, and operations continued until near the end of the planned run when it also plugged. Rather than prematurely end the run or replace the melter, it was decided to attempt to drill out the bottom nozzle. This effort was successful, and the processing run was completed in July 1991 using the second melter. Table 2 summarizes the total production data.

Currently a reduced staff of operations personnel are disassembling the second melter starting at the melter top and working toward the bottom. Waste is being broken/cut up and placed in drums subject to a 2 Sv (200 R) contact dose limit which also serves to limit heat generation. Following disassembly of the second melter, the first melter will be moved into the processing cell and disassembled.

Consideration is being given to process at PAMELA an additional 60 m³ of German waste in the 1996/1997 time period that would be trucked to the plant in 3.7 m³ shipments following installation of a new melter and waste receiving facility at PAMELA and a waste packaging facility at Karlsruhe.
Table 2. PAMELA Production Data
Date 05.09.1991

<table>
<thead>
<tr>
<th></th>
<th>LEWC-CAMPAIGN 1.10.85 - 12.6.86</th>
<th>HEWC-CAMPAIGN 1.10.86 - 05.09.91</th>
<th>TOTAL</th>
</tr>
</thead>
<tbody>
<tr>
<td>VITRIFIED WASTE SOLUTION (m³)</td>
<td>47.2</td>
<td>860.1</td>
<td>907.3</td>
</tr>
<tr>
<td>ALPHA-ACTIVITY IN THE FEED (Bq)</td>
<td>1.28 E15</td>
<td>2.26 E14</td>
<td>1.51 E15</td>
</tr>
<tr>
<td>BETA-ACTIVITY IN THE FEED (Bq)</td>
<td>2.78 E17</td>
<td>1.64 E17</td>
<td>4.42 E17</td>
</tr>
<tr>
<td>WASTE OXIDES (t)</td>
<td>7.67</td>
<td>88.94</td>
<td>96.61</td>
</tr>
<tr>
<td>GLASS PRODUCT (t)</td>
<td>77.8</td>
<td>411.7</td>
<td>489.5</td>
</tr>
<tr>
<td>CONTENT OF WASTE OXIDE (%)</td>
<td>9.9</td>
<td>21.60</td>
<td>-</td>
</tr>
<tr>
<td>CONTAINER FILLED</td>
<td>60 l</td>
<td>567 ··</td>
<td>934</td>
</tr>
<tr>
<td>150 l</td>
<td></td>
<td>1501</td>
<td></td>
</tr>
<tr>
<td>TIME AVAILABILITY (%)</td>
<td>88</td>
<td>93.16</td>
<td>-</td>
</tr>
<tr>
<td>EFFICIENCY (%)</td>
<td>69</td>
<td>96.58</td>
<td>-</td>
</tr>
<tr>
<td>EMISSION ALPHA-ACTIVITY (Bq)</td>
<td>4.4 E3</td>
<td>4.08 E3</td>
<td>8.5 E3</td>
</tr>
<tr>
<td>EMISSION BETA-ACTIVITY (Bq)</td>
<td>3.4 E6</td>
<td>6.02 E5</td>
<td>4.0 E6</td>
</tr>
<tr>
<td>CUMULATIVE DOSE RATE (mSv)</td>
<td>40</td>
<td>211</td>
<td>251</td>
</tr>
</tbody>
</table>

BP WAK

PAMELA

* inclusive 18 from active test operation
NUCLEAR RESEARCH CENTER (KfK) - KARLSRUHE, GERMANY

The Karlsruhe Nuclear Research Center (Kernforschungszentrum, KfK) is located approximately 5 miles north of the city of Karlsruhe and occupies a site of approximately 2.5 square kilometers. The center is organized as a private company under German law, whose shareholders include the Federal Republic of Germany (90%) and State of Baden-Württemberg (10%). Close relations are maintained with the Universities of Karlsruhe and Heidelberg. The main activities of KfK are in research and development in nuclear, environmental, and other promising technologies. Presentations and discussions were held on activities carried out in the Institute for Nuclear Waste Management (INE) headed by Professor Dr. J.-I. Kim. These included research on long-term safety assessment of nuclear waste disposal and tests for direct disposal of fuel, which are also supported by another German government agency, treatment of acidic intermediate level sodium-bearing liquid waste concentrate by drying, and development of the vitrification process using a liquid-fed ceramic melter. KfK’s operation of decontamination facilities were visited and discussed. Discussions were also held with KEWA, a German utility which has capabilities in nuclear fuel reprocessing, robotics, vitrification technology, and waste management engineering design services. KEWA is a subsidiary of DWK which is owned by the German utilities and was originally in charge of developing, designing and constructing a reprocessing plant for the German utilities.

Long-Term Safety Assessment of Nuclear Waste Disposal

The work includes material research on waste packages, near-field chemistry and barrier functions, far-field chemistry and barrier functions, and thermal and thermochemical processes in disposal sites. Material research on waste packages (vitrified components, spent fuels, and package materials) includes research in the corrosion process and surface reactions, radiation effects on near-field chemistry, reaction products and formation of second barriers, and long-term mineralization and migration behavior. Near-field chemistry and barrier functions include thermodynamics and kinetics of radionuclides and near-field materials under conditions of radiation, redox,
gas generation, complexation, colloid formation, and dissolution/remineralization, with a goal of providing a geochemical model of the near-field chemistry. Far-field chemistry and barrier functions include similar work in the geology surrounding a repository and does not depend on the waste form properties directly. Thermal and thermomechanical processes of a disposal system are evaluated based on the temperature effect, mechanical tension, disposal tunnel convergence, and permeability and porosity. The safety assessment of geological disposal includes combining the geochemical behavior in near/far-field aquifer systems with near/far-field transport processes to provide radionuclide transport.

KfK personnel are involved in the evaluation of U.S. performance assessment modeling and are familiar with the approaches used here. They also have expertise in waste form testing such as required for the ICPP Spent Fuel and HLW Management Program, including systems analysis. While these presentations were brief, further contact should be explored with the KfK personnel to determine possible collaboration and technical exchange.

Direct Disposal of Spent Fuel

The current German Atomic Energy Act stipulates reprocessing as the reference process for waste management of spent fuel in the Federal Republic of Germany (FRG) based on an obligation to recycle the fissile materials. However, spent fuel direct disposal studies have been under way since 1979. By 1984 a technical concept had been developed and a study comparing reprocessing and direct disposal completed. In January, 1985, it was decided that direct disposal should be developed to maturity for those cases in which reprocessing either is technically not feasible or economically not justified. The German utilities in 1989 decided to abandon German reprocessing efforts and to use French and/or British reprocessing capabilities in the near term,
pending maturity of the direct disposal technology. An amendment to the Atomic Energy Act is being pursued which would allow direct disposal when reprocessing either is technically not feasible or economically not justified.

The Gorleben salt dome in lower Saxony, Germany, has been selected as the reference site for a high level waste repository. A license application was submitted in May 1986 for a spent fuel conditioning and encapsulation pilot facility at Gorleben. A partial construction permit (building) was granted in January 1990 and a second partial permit (components) is expected in 1993 with hot operation planned for early 1996. The Lower Saxony Government has objected to construction of the facility, and this may delay the schedule. It is estimated that an expenditure of 500 Million DM will be required to qualify conditioning and direct disposal including the pilot facility. German utilities will spend roughly 400 Million DM, the Federal Ministry for Research and Technology (BMFT) 75 Million DM, and KfK 25 Million DM.

Two options are being pursued for direct spent fuel disposal - the Pollux cask for disposal in drifts, which is the reference concept, and the Pollux canister for disposal in 300-m deep boreholes, which is the backup. The Pollux canister is the same as the canister used for vitrified waste. Handling would require a shielded cask, and fuel would have to be cut for placement in the canister. Criticality was stated as not expected to be a problem in the salt because of the high concentration of chloride, a neutron absorber. The Pollux cask consists of a thick outer portion fabricated out of ductile iron and an inner high-strength steel portion, coated with Hastelloy C4 to minimize corrosion. It provides for its own shielding but requires special handling equipment due to its weight. The cask is considered to give adequate control of safeguards for the fissile materials in the spent fuel. German studies of various disposal options conclude that the cost differences between various concepts are mainly caused by changes in types and numbers of containers that accompany changes in emplacement concepts. Some further specifics about each option are given in Table 3.
Table 3. Comparison of Specifications for the Pollux Canister and Cask

<table>
<thead>
<tr>
<th></th>
<th>Pollux Canister</th>
<th>Pollux Cask</th>
</tr>
</thead>
<tbody>
<tr>
<td>Diameter</td>
<td>43 cm</td>
<td>150 cm</td>
</tr>
<tr>
<td>Length</td>
<td>134 cm</td>
<td>550 cm</td>
</tr>
<tr>
<td>Weight</td>
<td>1.2 tonne</td>
<td>65 tonne</td>
</tr>
<tr>
<td>Content</td>
<td>0.5 PWR assemblies</td>
<td>8 PWR assemblies a</td>
</tr>
<tr>
<td>Surface Dose</td>
<td>&lt;10^5 mSv/hr</td>
<td>&lt;0.2 mSv/hr</td>
</tr>
</tbody>
</table>

a Fuel arrangement has recently been optimized to hold 10 PWR assemblies.

Testing to date indicates that the cask plus repository rail car weighing 85 tonne can be handled in the repository shafts. On-site tests are under way in which two parallel drifts have been excavated at a depth of 800 m and three electrically heated dummy containers of Pollux dimensions and weight installed and backfilled with crushed salt. These in-situ tests indicate that temperatures can be predicted very well but discrepancies exist in the predicted and measured stress and strain fields. Tests started in late 1990 and will take three years for completion.

Testing is under way to assess the need for and effectiveness of barriers such as container material and fuel. The corrosion resistance of the Hastelloy C4 material, so far planned for corrosion protection for the Pollux cask, is influenced by the way in which the anti-corrosive coating is manufactured. Leaching studies with spent UO_2 fuel with a burn up of 52 GWd/t U show leaching behavior to depend on burnup and on the irradiation history. After six months roughly 1% of inventory of U, Pu, Am, Cs and Sr has been dissolved.

In 1993, tests will begin to evaluate the behavior of neutrons from disposal packages with emphasis on neutron back scattering in drifts. The date of implementation of direct disposal in the FRG will be determined by
commissioning of the Gorleben repository, which is not projected to occur till 2008. Underground exploration of the Gorleben salt dome is likely to be finished between 1995 and 1999.

**Drying of Intermediate-Level Waste Concentrate**

The intermediate-level liquid waste (ILLW) concentrate at KFK is an acidic, sodium-bearing waste obtained from decontamination and solvent washing operations. It contains fission products/actinides up to 1 Ci/l, nitric acid, sodium nitrates, corrosion products, and organic impurities, existing partly in two phases. Vacuum drying of the ILLW concentrate would result in a salt with a five-fold volume reduction compared to the concentrated ILLW and a ten-fold volume reduction compared to a cemented ILLW product. Currently, the German utilities are using a drying process for reactor wastes containing borates. Laboratory (1-1) and technical scale (200-1) tests were run with inactive simulant to determine if the drying process is feasible and does not result in an unsafe, explosive condition with some of the organic nitrates. To avoid NO_x generation and release, the acidic ILLW simulant was neutralized before heating under vacuum. During all operations (heating, drying, cooling), no significant temperature excursions were found, indicating that an explosive material is likely not formed. Splashing was found to occur during heating, but could be controlled with mechanical or chemical additives, but required fairly sensitive process control. In the 200-1 test, the best product with minimum splashing was obtained in 390 hr, which they believed could be reduced by 30% with better heating and off-gas controls.

Although such a drying process may appear to be applicable in principle to treating ICPP sodium-bearing waste, it likely is not a practical option because of the large waste volume of 6000 m^3_ and the significant scale-up required of the 200-1 batch operation for 390 hr per batch. In addition, a drying process for ICPP waste needs to take into consideration the impact of product composition on waste form requirements.
The basic operating data for PAMELA vitrification of Belgian HLW was presented earlier in this report. Discussions at KFK were continued on noble metal accumulation and proposed melter modifications to help overcome this problem. The primary fission product noble metals found in the waste included ruthenium, rhodium and palladium as oxides and alloys, including RuO$_2$ and Pd$_x$Te$_y$. The densities for these compounds are high, 7 g/cm$^3$ for RuO$_2$ and 12 g/cm$^3$ for Pd$_x$Te$_y$ and the particles agglomerate into RuO$_2$ needles and Pd$_x$Te$_y$ droplets. The morphology of the RuO$_2$ sediments changes with the formation of up to 200 $\mu$m needles, creating a non-newtonian liquid with increasing viscosity as a function of time. For glass enriched with 15 wt% RuO$_2$ and 4 wt% Pd, the electrical conductivity and viscosity each became 10 to 100 times higher compared to normal glass, resulting in the "short circuit" by the electrical current. As observed at PAMELA, there is relatively high viscosity and no apparent noble metal depletion from the melter due to glass pouring into canisters. An advanced melter, K6, was designed with steeply sloped sides and a bottom drain to allow for continued removal of noble metals during operation. Placement of electrodes was also modified to result in better heating control.

Physical and mathematical modeling of the modified melter was carried out, producing calculational details of the electrical power density, temperature and velocity fields. Flow studies are under way to verify behavior of noble metal sludge layers along inclined melter walls, material properties and necessary inclination.

Other vitrification projects currently under development at KFK include technology transfer to the People's Republic of China, including fabrication, testing, and delivery of a 70 l/hr nonradioactive melter for 1994-5 operation. A 1/10 scale Savannah River Site/Hanford melter was fabricated and tested in a cooperative venture to determine impact of noble metals on the U.S. melter design. While increased accumulation of noble metals was observed, no detrimental impacts on operation were noted.
The pilot-plant melters, including K6 with sloped sides, Chinese prototype, and SRS/Hanford melters, were toured. During the tour, the KfK engineer proposed another possible explanation for bottom pour nozzle plugging during the active PAMELA runs -- that increased current densities overheated the drain, causing drain localized melting. For the plugging of first active PAMELA melter, the increased localized current density could have been caused by the noble metal sediment and increased current density directed at the bottom drain. For the second active PAMELA melter, where noble metal concentrations were much lower, a possibly abnormally high alumina concentration in the waste solution might have prevented initial pouring. A subsequent attempt to increase in current density in order to overcome the pouring problem might have also inadvertently overheated the bottom drain, causing localized melting.

**Decontamination Facility**

The Center for Decontamination Operation (HDB) was toured, and a summary flowsheet is shown in Figure 5. This facility is used for decontamination of many KfK and offsite wastes, including all low and medium-level wastes which arise in the KfK area, collection, treatment, and controlling of all domestic sewage and chemical liquid effluents handled at KfK, and collection and treatment of all chemical hazardous wastes. Treatment techniques include evaporation, sewage treatment, drying, cementation, increased purification, sandblasting, electropolishing, compaction, incineration, and metal melting. Metal can be recycled without restriction if the contamination level is below 0.1 Bq/g and with restrictions that it would not experience daily human contact if the contamination level is below 1 Bq/g. For approximately 10,000 m³ of waste previously treated, 1,000 m³ of low heat generation waste product and 40 m³ of heat-generating waste product were produced.

**Discussions with KEWA Representative**

Discussions were held with K.-D. Kuhn representing a privately owned company specializing in service related engineering/technology of fuel reprocessing. KEWA is wholly owned by DWK, a company owned by the German
Structure of HDB-plants
utilities to develop nuclear fuel reprocessing and waste management. A pilot plant for reprocessing is located at KfK and was operated by WAK, another wholly owned subsidiary of DWK. KEWA is interested in providing services where needed in areas of technology, including results of experimental work of DWK and KfK and hot operation of the reprocessing pilot plant (WAK) and vitrification plant (PAMELA). Knowledge is also available from the design work performed for the projected German industrial reprocessing plant at Wackersdorf (WAW, now cancelled). The design of WAW was based in part on facilities at ICPP and has some similar characteristics. DWK has in the past also developed modern techniques for remote handling using cranes and TV and for remote cutting and welding of piping. Because of the fact that Germany has some of the most restrictive off-gas emission controls, KEWA'S expertise in this area was cited to be especially strong. Examples of past/current KEWA engineering services include the following:

- Design of off-gas treatment plant by KEWA/LURGI for JNFL/Hitachi (Japan).
- Experimental testing of iodine retention material for Hitachi (Japan).
- Design criteria for Ru retention in vitrification for IHI (Japan).
- Study on reprocessing site criteria for MITSUI (Japan).
- Study on feed clarification constituents for COGEMA (France).
- Study on cost of Kr-85 retention technology for BNFL (England).
- Establishment of PAMELA inactive melter mockup for BINE (China)/German engineering consortium.
The proposed steps in transferring technology include providing a free Know-How summary report of the candidate technology, followed by a contracted detailed synthesis report of the desired technology, if of interest. They proposed that they would give us an honest assessment of their abilities in a particular task if we have some interest. An example of a "Know-How" report was presented, and a listing of some of the technological areas of DWK expertise includes the following:

- Remote handling equipment for large hot cells
- Flowsheet investigation of technical scale extraction facilities
- Operation of the WAK evaporators
- Centrifuge or filter and technical data base for liquid feed clarification
- Off-gas cleaning and surveillance
- Improvement of solvent wash apparatus
- Solvent degradation; solvent removal from aqueous systems
- Treatment and recycle of contaminated material
- Incineration of burnable waste
- Scrapping and compaction of wastes
- Treatment of effluents
- HLLW Transportation
- HLLW Vitrification
- ILLW volume reduction
The La Hague complex is located 25 kilometers west of Cherbourg at the tip of the Cotentin peninsula on a site 3 kilometers long by 1 kilometer wide. It has a full-time staff of approximately 4,500 people. Since opening in 1966 the La Hague plant has reprocessed fuel from three reactor types: Gas-cooled, fast breeder and light water reactors. Since 1987 it has been dedicated exclusively to reprocessing light water reactor fuel. Its current spent fuel reprocessing capacity is 1200 tonnes per year with a projected increase in capacity to 1600 tons in 1994. The La Hague facility together with uranium mining at six sites, enrichment at two sites, and fuel fabrication at eight sites is operated by COGEMA which was incorporated in 1976 as a private industrial group whose stock is wholly owned by the French Atomic Energy Commission (CEA). COGEMA employs between 16,000 - 17,000 and had a revenue of 21.7 billion FF ($ 4.3 billion) in 1991. The visit to La Hague provided information on process flow at the facility and focused discussions on waste treatment and storage.
Process Flow

The reprocessing operations at La Hague are illustrated in Figure 6. Upon discharge from the reactor, spent fuel is stored for one year at the power station and shipped by rail or by boat to Cherbourg, then overland by rail to Valognes and La Hague. Since 1966 roughly 5,000 casks have been received and they are currently receiving 300 casks/year. Upon arrival at La Hague the casks are unloaded in either the Wet or Dry fuel unloading facility each with a capacity of 1000 tonne/yr, which is due to be increased to 1600 tonne/yr each by 1994. For wet unloading, the cask is prepared for unloading in water, placed in a pool for cooling and testing for damaged fuel while in the cask, following which the fuel is removed from the cask and placed in baskets for storage. In dry unloading, the cask is attached by mating the top of the cask to the floor of a shielded cell from underneath, the cask lid is removed remotely, each fuel assembly is then removed separately and placed in a cooling pit where damaged fuel is also detected, and after cooling is put into baskets for storage. In either case the fuel is stored in 9-m deep storage pools in 4 x 4 array containers for a minimum of 3 years prior to reprocessing. The dry unloading was felt to be the better of the two approaches as it requires less handling, internal cask cooling is not required, isolation of any broken fuel is readily accommodated by segregation of individual fuel elements in the dry unloading fuel cooling pit, and washing of the external cask is not required since only the top of the cask is in contact with the unloading cell. Available pool storage consists of four pools with a 10,000 tonne fuel load capacity that could be upgraded to 14,000 tonne. No attempt is made to consolidate and reduce the volume of a given number of fuel elements such as BNFL does with their AGR fuel, but COGEMA indicated that the technical capability was available to reduce stored fuel volume by 40%.

Following a minimum storage time of three years at La Hague (plus one year at the power station), the fuel is reprocessed in either the UP3 plant that began operation in 1990 with a capacity of 800 tonne/year for commercial fuel from foreign customers or the original UP2 plant for French utility’s fuel with a current capacity of 400 tonne/yr, which is being upgraded to
Figure 6. Reprocessing Operations at the La Hague Site.
UP2-800 with a capacity of 800 tonne/yr in 1994. In either case the fuel elements are crushed together and cut into 3 cm long cylindrical segments or rounds which are fed into a 90°C nitric acid bath continuous dissolver. The rotary dissolver is a paddle wheel arrangement which takes a group of fuel segments or rounds and moves them through the dissolver nitric acid bath and then discharges them after two hours, following dissolution of the rounds core material, to a rinse followed by encapsulation in concrete. An anti-floatation device is utilized to obtain the desired distribution of the zirconium fuel rounds and other cut up support plate components in the encapsulatary concrete. Future plans include compaction or volume reduction by melting.

The acid containing fission products, actinides, plutonium and uranium is processed through mixer-settler and pulsed column separation processes where the reusable uranium and plutonium are removed and separated/purified. Reusable uranium is shipped as a liquid uranyl nitrate solution to the enrichment plant in 120-l containers. The plutonium is converted to a dry plutonium oxide and placed in 3-kg cylindrical containers which in turn are packaged in groups of 4 or 5 for shipment to the fuel fabrication facility.

Raffinate from the separation process is concentrated and stored in 120-m³ tanks for a minimum of one year before processing through the vitrification plant (total time out of reactor is 5 years). Vitrification facilities at La Hague consist of two 3-line plants each line having a 30 kg/hr through-put capacity. Normally only two lines in each plant are operating with the third line being in maintenance or standby. See Figure 7 for the flowsheet of one of the AVH vitrification lines. After the one-year minimum storage period, the liquid waste is fed together with additives including sugar, sodium, calcium, and aluminium to a sloped rotating (20 rotations per minute) tube rotary kiln calciner with zoned resistant heating. Inlet temperature is approximately 650°C and outlet temperature is approximately 850°C. Calcine from the calciner and glass frit in a 22% to 78% ratio are continuously fed into an Inconel 601 oval cross section induction-heated melter whose outside skin temperature is maintained at approximately 1130°C with an inner molten glass temperature of 1080°C. Argon
Figure 7. French AVH Vitrification Process Flow Diagram.
sparge is used to agitate the molten mixture and to maintain an inert atmosphere to minimize corrosion. Roughly every 8 hours one-half of a waste canister volume (200 kg) is removed from the melter over a weir that is designed to maintain a 70 kg heel. Following receipt of two melter discharges, the filled glass canister is allowed to cool naturally after which a lid is welded on the canister. The canister is checked for contamination, decontaminated if necessary, and then moved to an interim storage area.

The current high-level waste glass storage area consists of 5 bays each with a 5 x 20 array of silos capable of storing canisters 9 deep. Canister heat is limited to 3.0 kW and forced air cooling is used to ensure that the glass centerline temperature is maintained below 500°C. The canisters will be stored in the facility for a minimum of 5 years, and this will reduce the heat loading below 2 kW to approximately 1.5 kW, after which the canisters will be moved to a storage area for French waste. Foreign waste glass will be shipped to the customer beginning as early as 1994. Additional storage capacity is being added which will become operational in 1994, and this will double the current La Hague capacity. Current capacity is approximately 30% full.

Focused Discussions

Alternative Technologies for Dispositioning of Spent Fuel

As in the case of BNFL at Sellafield, future orders by the utilities to reprocess spent fuel could depend on the economics of reprocessing versus direct disposal. When asked about their economic evaluation, COGEMA felt that while the reprocessing costs are well established, fuel disposal costs have not yet been determined, and a direct comparison cannot be made. However, they felt that their operation would be more environmentally responsible in that the plutonium is recycled, and only fission products are disposed in the glass at an overall volume reduction. Such an operation is consistent with non-nuclear industries, in which there are increasing requirements to minimize wastes by recycling and other means. As an example they stated that for every eight fuel assemblies (total of 4 tonnes uranium) disposed under the direct disposal option, only one fuel assembly plus two 150-l glass canisters are
produced as high-level waste for the reprocessing case with plutonium recycled in mixed-oxide fuel. While additional intermediate-level and low-level wastes are produced in the reprocessing/Pu recycle case, their goal is to reduce the intermediate-level waste to 1.5 m$^3$ per tonne of uranium.

**Waste Treatments and Emissions**

Waste treatment at La Hague consist of vitrification of high level waste (raffinate after separation of uranium and plutonium), concrete encapsulation of solid intermediate level waste (spent fuel rounds and fuel support structures), bituminization of intermediate level waste sludges resulting from cleanup of various process streams, and grouting of various technical waste residue associated with monitoring, control and process maintenance. A summary of their volumes of residues, current and projected, is provided in Table 4 below. All categories are considered, including non-surface and surface disposal.

<table>
<thead>
<tr>
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<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Fiss. Prod</td>
<td>HLW Glass</td>
<td>130</td>
<td>115</td>
<td>115</td>
<td>115</td>
</tr>
<tr>
<td>ILW Rounds</td>
<td>Concrete</td>
<td>600</td>
<td>600</td>
<td>&lt;600</td>
<td>150</td>
</tr>
<tr>
<td>ILW Sludge</td>
<td>Bituminization</td>
<td>630</td>
<td>450</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>ILW Residue</td>
<td>Grout</td>
<td>1700</td>
<td>200</td>
<td>200</td>
<td>&lt;200</td>
</tr>
<tr>
<td>TOTAL</td>
<td></td>
<td>3060</td>
<td>1365</td>
<td>&lt;915</td>
<td>&lt;465</td>
</tr>
<tr>
<td>LLW Near Surf.</td>
<td>Grout</td>
<td>3800</td>
<td>1400</td>
<td>1400</td>
<td>&lt;1400</td>
</tr>
</tbody>
</table>
The decreases in bituminization waste volume will be accomplished by process recycle and putting the sludge waste in glass, which will offset the planned reductions in vitrification glass waste due to improvements in process efficiency. Decreases in the rounds waste volume will be accomplished by crushing and/or melting before encapsulation.

Effluent emissions were also discussed in some detail. Kryton and iodine releases at La Hague have increased from 12,000 TBq in 1976 to 101,000 TBq in 1991 compared to an annual authorization of 480,000 TBq. An anomalous peak of 70,000 TBq occurred in 1985 due to an accident. Alpha-beta aerosol release decreased from 3.3 GBq in 1976 to 0.026 GBq in 1991 compared to annual authorization of 74 GBq. A peak of 70 GBq occurred in 85 due to an accident. Halogen (including iodine) releases increased from 12 GBq in 1976 to the 25 to 28 GBq range in the 1989 to 1991 time period. The annual authorization is 110 GBq. Discharge to the sea of α emitters decreased from 0.37 TBq in 1976 to 0.153 TBq in 1991 compared to an annual authorization of 1.7 TBq. Also β emitters discharged overall decreased from 730 TBq in 1976 to 116 TBq in 1991 compared to an annual authorization of 1700 TBq. The plant did experience peaks of 1110 TBq in the 1982 - 1987 time frame. Average occupational radiation exposure of overall plant personnel appears to set an industrial benchmark of 0.50 mSv/yr or 50 mrem/yr, and exposure to maintenance workers is somewhat higher.

Vitrification Operations

Discussions with Vitrification UP2 (R7) personnel indicated that they were achieving an average melter life of 1650 hours and were hoping to raise that to 2000 hours in 1993 and 5000 hours in five years. They indicated that the main cause of failure was corrosion and that the failure phenomenon was similar to that described by BNFL (pin-hole failure, loss of vacuum, followed by cracking). They attributed the longer average melter life compared to the Sellafield data to careful temperature control and more operating experience. It was noted that a melter in the UP3 (T7) facility has run for 2600 hours and that this melter was utilizing a third generation temperature control system.
They indicated that investigation of alternate materials to provide increased corrosion resistance has been unsuccessful to date in obtaining a material superior to Inconel 601.

When asked about melter changeout time they indicated that this was routinely accomplished in 2 days including restart. They attributed their ability to perform a changeout in 2 days compared to the 6 day to 6 month time period reported by BNFL to the training/experience of their personnel and the organizational management structure. Also it was noted that their plants have three lines versus 2 lines at Sellafield and one of the French three liners is always in maintenance or standby. They indicated that they had no specific difficulties associated with reliability and maintainability of auxiliary equipment (cranes, manipulators, etc.) similar to that experienced by the British. They noted that their UP2 (R7) plant had produced 450 canisters during the past year and 1350 canisters during the last three years.

When asked about additives to their waste they indicated that sugar was added for NOx and ruthenium retention, lithium, aluminum, calcium and sodium for cesium and strontium retention in the glass. They identified that increased melter life was their most important process improvement from an operations point of view. Their objective is to achieve a 1.00-hr melter life and feel that the best possibility for achieving this is through changes in the melter geometry and materials.
Marcoule, which is located in southern France, near Avignon, is a defense site with production/power reactors (gas cooled, graphite moderated), breeder prototype PHENIX reactor, the first reprocessing plant (UPI), and the first waste glass production facility (AVM) starting in 1978. The vitrification process was demonstrated at Marcoule at a capacity of 15 kg/hr using a cylindrical metal melter and was the basis of the AVH process design at La Hague, where each vitrification line uses an oval-shaped metal melter with a 25-30 kg/hr processing capacity. Both CEA and COGEMA are involved at Marcoule, with COGEMA as the operator of the reprocessing and vitrification plants. Currently the center employs approximately 2300 people of which roughly 80% are COGEMA employees and 20% are CEA employees.

The purpose of the visit was to discuss ongoing work in the Fuel Reprocessing Waste Management (DPR) division of CEA with a focus on new developments in melter technology as related to vitrification and metal melting and other topics. The CEA operates laboratory and pilot-plant facilities from hot cell laboratory testing, waste from characterization, waste component separations tests, cold crucible induction heating metal and glass melters, and a full-scale AVH prototype nonradioactive melter for support of flowsheet development at AVH in La Hague. For example, tests were in progress at the prototype in vitrifying a high molybdenum (Mo) and ruthenium (Ru) content waste stream with compositions expected from a future AVH reprocessing flowsheet while the pilot plant was being toured. A new hot-cell facility, which will be used for French and other European Commission member countries' testing, is ready for start-up after permitting has been completed.

Overview of Current Fuel Reprocessing Waste Management (DPR) Division Activities

An overview of current DPR division activities was described to include:

- Reprocessing R&D
- Support of Melox (Mixed oxide fuel fabrication) Plant
- Head-end Process R&D
- Plutonium Extraction
- Vitrification
- Liquid Effluent Treatment
- Computer Assisted Control
- Advanced Processing
- Waste Treatment
- Decontamination
- Technology Spin Off (such as use of solvent extraction for perfume purification)

Available laboratory-scale facilities include hot cells and radioactive laboratories. Pilot-scale facilities include hot cells, α-waste glove boxes, and up to full-scale non-radioactive mock-ups.

Reprocessing R&D on head-end processes includes fuel dissolution, fuel solubility, solution clarification, fines characterization, gaseous effluent treatment, and hulls characterization and treatment. Hulls melting tests are carried out at Marcoule and compaction tests are done elsewhere. Solvent extraction tests include plutonium extraction, behavior of chemical species, process and flowsheet design and various extraction methods.

Vitrification R&D includes material studies of the glass and immobilized fission products, characterization, and long-term waste form behavior. Liquid effluent waste treatment improvements are under way to reduce waste discharge by recycle and to improve the quality of the disposal package. The reprocessing plants at Marcoule and La Hague plan to stop generating bitumen out of the intermediate-level waste (including Na-bearing waste) by decreasing use of sodium (e.g. as only for final solvent wash) and increased recycle of various process chemicals. The radioactive components of the intermediate-level waste will be incorporated in glass. There is some interest in CEA in "advanced reprocessing" R&D for actinide separation and transmutation as a method for reducing waste volume and α-waste inventory in the repository. New solvents, including diamides, were being tested, and the goal is to remove Pu, Np, Am, Cm, Tc, and Cs as long-lived isotopes. This
approach has the French acronym SPIN, and the test facilities are currently being transferred from the Fontenay aux Roses facility in Paris to Marcoule in order to remove all radioactive α-waste testing out of Paris.

**Melter Technology**

Key features associated with the operational life of the AVM melter were described as:

- Good temperature control
- Use of an inert argon gas atmosphere above the glass melt in the melter
- Agitation to ensure a homogeneous mixture within the melter
- Good melter material grain size control and weld quality
- Good heat transfer characteristics within the melter

It was identified that investigation of alternate melter materials such as Inconel 690 did not produce any significant improvement over Inconel 601 to warrant a change. It was also identified that no basic changes in melter geometry were being pursued; however it was indicated the vanes within the melter are being introduced to improve the heat transfer characteristics within the molten glass. It was also identified during discussions that lithium is added to the glass frit mainly for cesium and strontium retention and sugar to the waste mainly for ruthenium retention but also to improve calcination. Other additives to the waste are aluminum nitrates and calcium nitrate to improve calcination. Glass frit is fed at 19.8 kg/hr and waste at 50 L/hr.

**Cold Crucible Melter**

A new glass and metal melting furnace concept, the cold crucible melter, using induction heating directly coupled to the glass or metal melt and cooled melter walls using internal water flow was described. The primary difference between this concept compared with the induction heated furnaces used in the existing glass process at AVM in Marcoule and AVH in La Hague and Sellafield
is that in the existing melters all of the induction heat is directly coupled to the metal melter's wall. The heat is then transferred thermally from the melter wall inside to the glass, requiring that the melter wall material must withstand extreme temperatures and resulting in limited melter operating lifetimes of 1200 to 2000 hours. The cylindrical cold crucible melter is fabricated out of curved metal axial segments which are separated by narrow gaps and are water cooled by internal channels. The gap between segments allows the induction current to penetrate to the glass inside the melter. A small amount of molten glass must be generated initially by an external means, such as plasma arc or microwave heating, in order that a current can be induced in the molten glass resulting in Joule heating. Solid glass does not conduct an appreciable electrical current and must first be melted externally before Joule heating can begin. Material is continually extracted from the lower melter elevation and new material (frit plus calcine) is introduced at the upper melter elevation for melting as the lower material is removed. By keeping the melter walls cooled, a solid glass layer remains on the walls and protects the metal from corrosive molten glass. The glass layer can be separated completely when the melter is cooled because of thermal expansion differences. The same heating concept also holds for metal inside the melter, such as for added fuel hulls, where the induction current melts the hulls but not the water-cooled melter walls. This cold crucible concept was invented under a 1925 German patent, and additional work in metal melting was done about 20 years ago by the U.S. Bureau of Mines at Albany, OR.

The CEA is interested in using the cold crucible initially in developing a higher throughput vitrification process for low-level and intermediate-level waste as well as for possible replacement of the AVH melters in the future. Because of the cooled melter walls, the melting temperature of the glass is not limited to 1130°C, but can be increased to 1400°C, which would result in a larger throughput. A 25-cm diameter by 70-cm long prototype melter has been operated with a 50 to 100 kg/hr glass melting rate. (The AVH capacity per line is 25-30 kg/hr). A 1-m diameter melter is expected to produce the desired production rate of 200 kW h. The power level required for the cold crucible approach is estimated as 2 kW h per kg glass produced, which is higher than required for a Joule-heated ceramic melter. Work is underway to
develop the cold crucible melter to process intermediate-level waste for Czechoslovakia.

The French are also investigating use of the cold crucible melter concept for melting into ingots spent fuel hulls or rounds produced from fuel dissolution. Although the current method for immobilizing the zircaloy cladding spent hulls remaining after fuel dissolution produces a stable concrete monolith, the immobilized volume and weight are significantly increased by more than a factor of 2 and 5, respectively, compared to the original hulls. A similar furnace was constructed of curved metal segments for the metal melting as for the glass tests, with the same induction heating technology run at higher temperatures approaching 2000°C. Some salt at 3 wt% must be added as a high temperature flux (CaF$_2$ for stainless steel, CaF$_2$ and BaF$_2$ for zircaloy) and for potentially removing some of the radionuclides as a slag. The metal and salt mixture is added to a furnace with a cooled movable bottom plate, and heating is induced by the induction coils. As the hulls melt, the bottom plate is slowly lowered, extruding an ingot of the hull metal. Hulls have been melted in a cold crucible non-radioactive laboratory-scale unit (13 cm diameter by 50 cm high) and full-scale prototype of 20 cm diameter by 1 m high. Hot laboratory-scale tests were done at 6 cm diameter by 20 cm high. A full-scale (20 cm x 1 m) hot test system is under construction. Tests using radioactive stainless steel and zircaloy hulls resulted in a decontamination factor for α-radionuclides of 1000 for stainless steel but much lower for zircaloy because zirconium effectively reduces plutonium and neptunium oxides and incorporates them in the metal rather than the slag. Table 5 gives the results for stainless steel hulls melting.
Table 5. Percent of Original Radioactivity Before and Remaining After Melting of Stainless-Steel Hulls

<table>
<thead>
<tr>
<th></th>
<th>in input</th>
<th>in slag</th>
<th>in off gas</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cs-137</td>
<td>&lt; 0.3</td>
<td>39.6</td>
<td>60.2</td>
</tr>
<tr>
<td>Co-60</td>
<td>99.97</td>
<td>0.02</td>
<td>&lt; 0.01</td>
</tr>
<tr>
<td>Ru-101</td>
<td>95.2</td>
<td>4.4</td>
<td>0.4</td>
</tr>
<tr>
<td>β emitters</td>
<td>85</td>
<td>10</td>
<td>5</td>
</tr>
<tr>
<td>α emitters</td>
<td>&lt;0.1</td>
<td>99.5</td>
<td>0.4</td>
</tr>
</tbody>
</table>

Thus, while it may be possible to remove sufficient α-contamination from stainless steel hulls to produce a non TRU waste, it is not possible for zircaloy hulls. In both stainless steel and zircaloy metals other contaminants such as cobalt (α emitter) were not removed from the metal melt, but were incorporated in the ingots.

Incineration Development

The CEA has developed at Marcoule an incinerator for high α-contaminated waste, such as expected from the mixed-oxide fuel fabrication plant under construction at La Hague.

The incinerable waste includes:

- Kleenex 5%
- Cotton wool 5%
- Latex 17%
- Neoprene 17%
- Polyethylene 8%
- Polyvinyl chloride 48%

The incinerator consists of two rotary kiln calciners in series, with the first operated under oxygen-free conditions to pyrolyze the waste at 550°C. The resulting product is a pitch-like material that is fed directly into the second calciner, which is operated at 900°C under aerobic conditions to
destroy the carbonaceous material and produce an ash. The ash is fed into an induction heated melter to immobilize the TRU waste in a glass. This technology results in less than 1 wt% of the plutonium carried into the off-gas system. The off-gas system consists of a 1100°C post-combustor followed by a dry cooler, prefilters to remove most of the particulates, and high-efficiency particulate filters (HEPA) to remove small particulates, followed by water scrubbing to remove chlorides in a radionuclide-free system. CEA has developed a non-radioactive 4 kg/hr process, and a radioactive 7 kg/hr plant will be built at Dijon for CEA. It is felt that they could go to 20 kg/hr without additional scale-up tests. The waste is initially processed through a waste segregation/purification system consisting of an x-ray detector, sorting zone crusher and pneumatic screening machine which air lifts the lighter waste. The total DF for the off-gas system is $2 \times 10^8$ and the weight reduction factor, defined as ratio weights of waste to ash, is 38. A combustion burn efficiency of 99.98% is achieved with 7 ppm CO and 4.5% CO$_2$ released. Treatment options being considered for incinerator ash and filter residues include:

**Ash**
- Electrolytic dissolving to recover Pu
- Vitrification
- Embedding in cement

**Prefiltration Dust**
- Water dissolving and filtration
- Electrolyses
- Purification with ZnCl$_2$ distillation

**Pilot Plant Tour**

The non-radioactive pilot plant was toured, and ongoing cold crucible test systems were seen, including ingots of metal produced by the small and full scale systems. A glass pour from the full scale AVH prototype was
observed for testing a new flowsheet waste composition containing molybdenum and ruthenium. The incinerator mock-up was also seen. The facilities and ongoing work appeared to meet the development needs for their R&D effort.

**Waste Form Characterization and Long-Term Testing**

The compositions of French waste feed, frit, and resulting glass product are shown in Table 6. The glass has been characterized for a number of properties such as shown in Table 7, including properties during production, leach rates, mechanical properties, thermal properties and radiation resistance. Long-term behavior of their glass has been determined for actinide leach rates, radiation effects, and alteration under disposal conditions, as shown in Table 8. Extensive testing is continuing in these areas for the different glass compositions prepared or expected for French wastes.

**Metal Melting/Recycling**

A handout was given to us on metal processing for the dismantling of early reactors and gaseous diffusion plants at Marcoule. Metal melting is being tested for steel and aluminum using a test facility located in one of the reactor buildings. The purpose is to reduce the volume, homogenize the residual contamination within the material, and recycle material as shielding or containers for higher activity materials. An electric arc furnace of nominal 15 ton capacity was chosen for steel tests, and trial runs producing a total of 3 tons have been completed as of June 1991. The handout and name of the responsible engineer is available for further review.
Table 6
Composition of French AVH HLW, Frit, and Glass

<table>
<thead>
<tr>
<th>Fission Product Solution, g/l</th>
<th>Frit, wt%</th>
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<tbody>
<tr>
<td>Na$_2$O</td>
<td>SiO$_2$</td>
</tr>
<tr>
<td>19.82</td>
<td>58.84</td>
</tr>
<tr>
<td>Al$_2$O$_3$</td>
<td>Al$_2$O$_3$</td>
</tr>
<tr>
<td>7.14</td>
<td>4.28</td>
</tr>
<tr>
<td>(Fp+Act)Ox</td>
<td>B$_2$O$_3$</td>
</tr>
<tr>
<td>54.0</td>
<td>18.15</td>
</tr>
<tr>
<td>Fe$_2$O$_3$</td>
<td>Na$_2$O</td>
</tr>
<tr>
<td>12.38</td>
<td>7.0</td>
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<tr>
<td>MO</td>
<td>CaO</td>
</tr>
<tr>
<td>1.85</td>
<td>5.23</td>
</tr>
<tr>
<td>Cr$_2$O$_3$</td>
<td>ZrO$_2$</td>
</tr>
<tr>
<td>2.26</td>
<td>0.70</td>
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<tr>
<td>P$_2$O$_5$</td>
<td>Li$_2$O</td>
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<tr>
<td>1.26</td>
<td>2.56</td>
</tr>
<tr>
<td>ZrO$_2$</td>
<td>ZnO</td>
</tr>
<tr>
<td>2.04</td>
<td>3.24</td>
</tr>
</tbody>
</table>

Calcination

Vitrification

<table>
<thead>
<tr>
<th>Glass Composition, wt%</th>
</tr>
</thead>
<tbody>
<tr>
<td>SiO$_2$</td>
</tr>
<tr>
<td>B$_2$O$_3$</td>
</tr>
<tr>
<td>Al$_2$O$_3$</td>
</tr>
<tr>
<td>Na$_2$O</td>
</tr>
<tr>
<td>CaO</td>
</tr>
<tr>
<td>Fe$_2$O$_3$</td>
</tr>
<tr>
<td>NiO</td>
</tr>
<tr>
<td>Cr$_2$O$_3$</td>
</tr>
<tr>
<td>P$_2$O$_5$</td>
</tr>
<tr>
<td>ZrO$_2$ (Filings)</td>
</tr>
<tr>
<td>Li$_2$O</td>
</tr>
<tr>
<td>ZnO</td>
</tr>
<tr>
<td>(Fp+Act)Ox</td>
</tr>
</tbody>
</table>
Table 7. GLASS INVESTIGATION CHARACTERIZATION STEPS

DETERMINATION OF PROPERTIES DIRECTED TOWARDS:

- FABRICATION
  - Viscosity
  - Volatilization during melting
  - Corrosiveness as regard to the process equipment
  - Expansion Coefficient
  - Specific gravity
  - Homogeneity
  - α and β emitters leach rates

- INTERIM STORAGE AND HANDLING
  - Mechanical properties
    - Young's Modulus
    - Fracture toughness
    - Rupture load modulus
    - Drop resistance (impact)
    - Microhardness

  - Thermal properties
    - Devitrification rate at various temperatures
    - Emissivity
    - Devitrification between 400 and 800°C

  - Radiation resistance
    - β irradiation effect

  - Thermal conductivity
### Table 8. GLASS TESTING FOR LONG-TERM BEHAVIOR

- Actinides leach rates
- Radiation effect
  - α irradiation effect (He buildup, devitrification Wigner effect)
- Basic mechanisms
  - Solubility limit in various ground water
  - Hydrothermal leaching (up to 300°C and 500 bar)
  - Specific behavior of Np-237, Pu-238, -239, Am-241, Tc-99
  - Surface layer behavior
- Alteration under disposal conditions
  - Static leaching (environmental materials under 90°C)
  - Semi-integral leaching (Redox effect, CO₂ effect)
  - Integral leaching (effect of materials fracturation, crystallization, pressure, etc.)

**Embedding of Alumina in a Glass Matrix**

A one-page handout was given to us of lab scale work completed in 1973 on encapsulating 1.4 mm alumina particles in a silicate-low-phosphate glass matrix. The best glass composition in wt.% included SiO₂ - 25.7; Al₂O₃ - 26.2; Na₂O - 14.3; B₂O₃ - 24.4, and P₂O₅ - 9.4. The results were published in a report, CEA R-4444.
Discussions were held with SGN personnel at their Paris suburban headquarters in Saint-Quentin. SGN is owned by CEA and COGEMA and provides a number of areas of technical support for the French nuclear activities. The company had its beginnings in 1952 as a department in the Compagnie de Saint-Gobain responsible for the design and construction of nuclear fuel reprocessing plants, forming Saint-Gobaine Nucleaire in 1960. After further diversification in 1964 and purchase of the majority of its shares by COGEMA in 1977, SGN is now called Society General pour les Techniques Nouvelles. Since 1980 SGN was the prime contractor for design and construction of the reprocessing plants at La Hague and has been expanding its export business. In 1989 the SGN Group and Eurosys Network were formed, including an association of SGN with many of its previous subcontractors to improve its flexibility. NUMATECH in the USA is a subsidiary company owned by SGN and COGEMA which is the prime coordinator of all SGN and COGEMA interaction with USA clients. Included as part of the Eurosys network for the SGN group are companies dealing with consulting, engineering, information technology, documentation and information, mechanical engineering and maintenance, and testing/technical assistance. It was suggested that if WINCO/DOE has identified some specific needs, the Eurosys Consultants could be contracted to pull together the specific expertise of the entire Eurosys Network of the SGN Group for possible technical solutions. Specific contracts to complete the task could then be issued to the appropriate subsidiaries with the necessary expertise/capabilities.

In 1991 the SGN Group had total sales of 3.0 Million French francs, 2,395 employees, and total available workforce through the Eurosys Network of 5,100 providing 6.3 Million hours of engineering and technical services. In France, SGN developed the design and constructed the La Hague processing plants, including vitrification facilities. Areas of service provided to outside organizations include LLW facility technology transfer to UK, design of a fuel shearing machine for Barnwell Nuclear Services, pneumatic sample transfer system to WWNS, technology for a fast drilling and analysis system to Hanford, and support on the Red Team and Readiness Review at SRS.
Some of the current areas of development at SGN include radionuclide separation techniques for Cs, Sr, Tc, and the water cooled metallic furnace (cold crucible furnace undergoing tests at Marcoule) with a potential capacity of 200 kg/hr glass, melt temperature up to 1500°C, and prolonged furnace lifetime. Potential applications include vitrification of medium-level liquid effluents, consolidation of zircaloy hulls, and vitrification of non-nuclear waste materials like asbestos.

A furnace using microwave heating is also under development and testing at another CEA site, in Cadarache near Aix-En-Provence. The cold crucible furnace using electric induction heating is considered most useful for continuous operations, while the microwave furnace would be best used for batch operations, such as melting batches of incinerator ash. Planned test systems include a 30 kg/hr pilot scale microwave melter at Marcoule and 60 l/hr pilot plant cold crucible melter for Czechoslovakian medium level waste as part of a cooperation agreement.

Key items of the vitrification process design were described, including special jumpers, pumps and valves. All pumps and valves have permanent enclosures located at the ceiling of a cell, with access to the pump or valve body from the maintenance cell directly above the operating cell via cell plugs. A removal-replacement system is located inside a shielded cask in the maintenance cell and can be positioned so as to carry out the operation. These systems are common to all French plants. Additional design features include a homogeneous metering system of a feed containing 10-30 wt% suspended solids and isolation of glass canister in a separate cell from the melter cell.

A brief overview of the waste and plant equipment qualification process was given. In facility qualification work, the objective is to demonstrate that the properties of the full-scale product are similar to the test samples and don’t change over time. Their methodology to qualify the facility include inactive operations using their actual procedures. The product qualification is controlled primarily by operating envelopes of the control system, with methods for detecting deviation in operations and counter measures for
restoring normal operation. Since they did not present the detailed qualification criteria, a request was made and agreement received that any nonproprietary criteria be provided to us of HLW glass, medium waste bitumen/cement, and LLW products.

The SGN capabilities appear to be thorough in the development of vitrification process and they seemed very eager to provide support for the new ICPP Spent Fuel and Waste Management Development programs. It is recommended that WINCO continue to explore areas of potential French support after a more detailed scope has been identified.
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FOLLOW-ON ACTIONS

• BNFL will forward available information on glass quality as a function of residence time in the melter.

• BNFL will furnish any available non proprietary information on specification of waste characteristics, vitrified waste glass characteristics, vitrification process additives, and wet scrubbing efficiency for NOx absorption.

• BNFL will furnish available information on influence of grouting on material solubility when exposed to liquid after grouting.

• BNFL to forward a package of information portraying their remote capabilities.

• WINCO to contact Malcolm Saunders of the UK concerning British decontamination development efforts.

• BNFL to forward a package of information portraying their Decontamination/Decommissioning capabilities.

• WINCO to continue to follow German effort on direct disposal of spent fuel.

• WINCO to continue to follow French and German Vitrification melter development efforts.

• WINCO to follow French efforts on cold crucible melter concept for applicability.

• WINCO to provide French with available information in pyrochemical processing.

• WINCO to investigate potential modifications of the German Intermediate Level Liquid Waste Drying Technology for use in processing sodium waste.

• SGN to forward available waste acceptance specifications for glass, cement, and bitumen
END
4/20/94
FILE
DATE