Qualification of a Vitrified High Level Waste Product to Support Used Nuclear Fuel Recycling in the US - 9474

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ABSTRACT

As part of the Department of Energy (DOE) Global Nuclear Energy Partnership (GNEP), AREVA formed the International Nuclear Recycling Alliance (INRA) consisting of recognized world-leading companies in the area of used nuclear fuel (UNF) recycling,. The INRA team, consisting of AREVA, Mitsubishi Heavy Industries (MHI), Japan Nuclear Fuel Ltd (JNFL), Batelle Memorial Institute (BMI), URS Washington Division and Babcock and Wilcox (B&W), prepared a pre-conceptual design for an upgradeable engineering-scale recycling plant with a nominal through put of 800 tHM/y. The pre-conceptual design of this leading-edge facility was based upon the extensive experience of the INRA team in recycling plant design and real world "lessons learned" from actually building, commissioning, and operating recycling facilities in both France and Japan. The conceptual flowsheet, based upon the COEX™ separations process, separates the useful products for recycling into new fuel and sentences all the remaining fission products and minor actinides (MA) to the high level waste, (HLW) for vitrification. The proposed vitrified waste product will be similar to that currently produced in recycling plants in France. This wasteform has been qualified in France by conducting extensive studies and demonstrations. In the US, the qualification of vitrified glass products has been conducted by the US National Laboratories for the Defence Waste Processing Facility (DWPF), the West Valley Demonstration Plant (WVDP), and the Waste Treatment Plant (WTP). The vitrified waste product produced by recycling is sufficiently different from these current wasteforms to warrant additional trials and studies.

In this paper we review the differences in the vitrified waste forms previously qualified in the US with that produced from recycling of UNF in France. The lessons learned from qualifying a vitrified waste form in Europe is compared to the current US process for vitrified waste qualification including waste forms, waste loading, and long term alteration models.

INTRODUCTION

In the perspective of future needs outlined by many international and national organizations, it is expected that nuclear energy will become a significant contributor to the world's energy mix in the future.

According to the recently published DOE/EIA report" *International Energy Outlook 2008*" (IEO 2008) [1], over the next 25 years, the world will become increasingly dependent on electricity to meet its energy needs. Electricity generation is expected to increase at an annual average of 2.6% through 2030 according to the *IEO 2008* reference case. Of this, the share of nuclear power is projected to increase from 2.6 TWh in 2005 to 3.8 TWh in 2030, as concerns about increasing fossil fuel prices, energy security and greenhouse emissions rise. The conservative growth projections considered by the INRA team in its proposal is compatible with these data, as shown in Fig. 1 below.

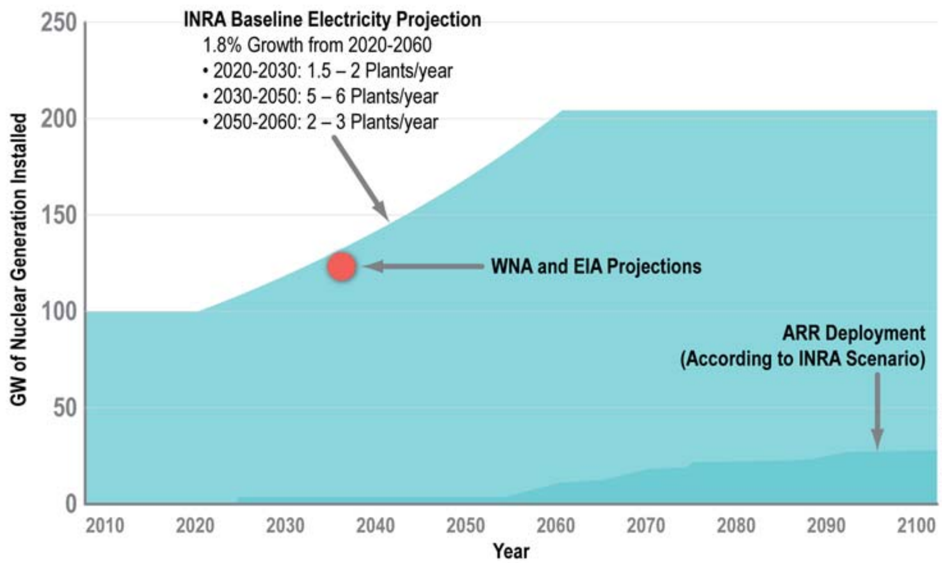

Figure 1: Projection for nuclear energy capacity growth during the 21 st century

In the US, the Energy Policy Act of 2005 recognizes the need for new electricity generating power plants and contains provisions to encourage the construction of new nuclear power plants. In the IEA 2008 reference case, 17 GW of new nuclear capacity are projected to come online by 2030. Such an expansion of nuclear power will require a complete, mature, and sustainable nuclear fuel cycle, that ensures safety for the public, enhanced non-proliferation, efficient use of resources, and minimal impact from waste management and disposal.

The Global Nuclear Energy Partnership (GNEP), initiative has been launched to provide a framework for both international and domestic use of nuclear power to reduce the risks associated with nuclear proliferation and the impacts associated with waste disposal. Internationally, GNEP comprises a partnership of countries working toward establishing international structures intended to prevent the uncontrolled spread of nuclear technologies and materials. Domestically, the Advanced Fuel Cycle Initiative (AFCI) program focuses on options for changing the U.S. fuel cycle to reduce the (HLW) radiotoxicity, volume and heat and supports the greater international GNEP vision. Recently, the DOE has published a Programmatic Environmental Impact Statement (PEIS) [2] which analyzes several alternatives for accomplishing the GNEP objectives. In its introduction, this document establishes that the DOE's preferred approach would be a change to a closed fuel cycle, although it evaluates several alternative pathways.

In 2007, DOE requested input from the nuclear industry regarding approaches to close the fuel cycle. In April 2008, DOE released a funding opportunity announcement (FOA) soliciting the nuclear supplier industry to explore the technical, financial and operational viability of a commercial used nuclear fuel management initiative, including a recycling facility – the Consolidated Fuel Treatment Centre (CFTC), and Advanced Recycling Reactor (ARR), as a complement to the geologic repository. DOE requested applications from the industry on endeavours to explore the technical and business parameters by providing studies in the main areas of business planning, technology development roadmaps and conceptual design studies. In response to DOE's request, AREVA, Mitsubishi Heavy Industries (MHI), Japan Nuclear Fuel Ltd (JNFL), URS Washington Division, Babcock and Wilcox Technical Services, and Battelle Memorial Institute formed the International Nuclear Recycling Alliance (INRA) to explore the commercial viability of used nuclear fuel recycling and advanced recycling reactors in the US. INRA combines the experience of the operator of the world's largest nuclear fuel recycling complex (AREVA), of the world leader in advanced fast reactors (MHI), of the owner and operator of the Rokkasho Reprocessing plant (JNFL), a world leader in technology and innovation (Battelle Memorial Institute), an expert in the protection of US nuclear assets; (Babcock and Wilcox Technical Services) and one of the world's leading engineering, procurement, and construction contractors (URS Washington division).

One important aspect of closing the fuel cycle is to determine an integrated waste management strategy for all the wasteforms produced by the facilities and, most importantly, for the HLW. The conceptual flowsheet for the CFTC proposes to separate the high level waste, (HLW) from the recyclable material and vitrify it, together with the minor actinides, in a glass matrix similar to that currently produced in recycling plants in France. The R7T7 vitrified wasteform currently produced at La Hague has been extensively studied and fully qualified for acceptance by AREVA's French as well as foreign customers and Safety Authorities. It is one of the major reference wasteforms for commercial HLW worldwide. The current presentation describes how this reference wasteform could be integrated into a US acceptance scheme, by making use of the experience already gained in qualifying Defence HLW at Savannah River Site (DWPF) and Hanford (WTP) and some commercial HLW from West-Valley, and adapting this experience to the nature, composition, and properties of the proposed wasteform.

PROPOSED APPROACH

During the initial phase of the contract with the Department of Energy (DOE), the INRA team considered several facilities of different sizes and determined that an upgradeable engineering-scale recycling plant with a nominal through put of 800 metric tons of heavy metal per year was best suited to the current market conditions in the US. This state-of-the-art facility is based on the team's extensive experience in recycling plant design and real world "lessons learned" from actually designing, building, commissioning, and operating recycling facilities in both France and Japan.

The first 800 tHM/y engineering facility that the INRA team will design, construct and operate will utilize the COEX™ process to recover uranium and plutonium as a mixed stream that will be used in the fabrication of new fuel for light water reactors, (LWR's). However it should be noted that AREVA believes it will also be possible to separate out the Neptunium, (Np) with the Pu by adjusting the process conditions within the plant. This separation of Np will initially only be conducted for transmutation fuel where U, Pu and Np will be co-precipitated. The initial 800 tHM/y facility will sentence all the MA and fission products to the HLW glass. In conjunction with the engineering efforts conducted by INRA, it is hoped that the national laboratories through the advanced separations campaign run through the Idaho National Laboratory will develop advanced processes for the separation of the minor actinide Americium, (Am) and the fabrication of Am Targets. Once this process has been developed by the laboratory the INRA team will work with the National Laboratory to industrialize the process and deploy it into the initial 800tHM/y facility by incorporating the technology in an annex to the main plant. This advanced process will then be used to separate Am which will be formed into targets for destruction in the Advanced Recycling Reactor's, (ARR's). Future commercial sized recycling facilities could deploy this advanced MA separations process if it is shown to be successful in the initial plant. Figures 2 and 3 below describe the proposed phased approach and the general flowsheet for the first CFTC facility. The reference fuel for the design of this facility is PWR fuel with a burnup of 50 GWd/MTHM.

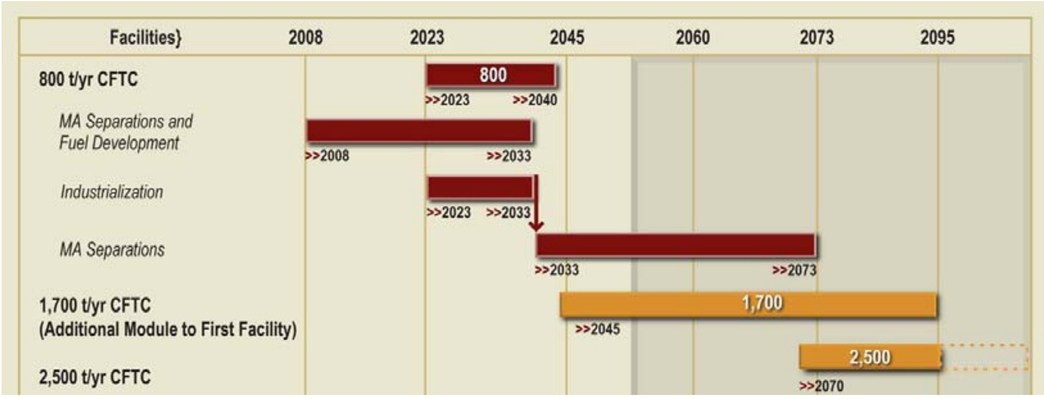

Figure 2 : proposed phased approach for implementing a closed fuels cycle in the US.

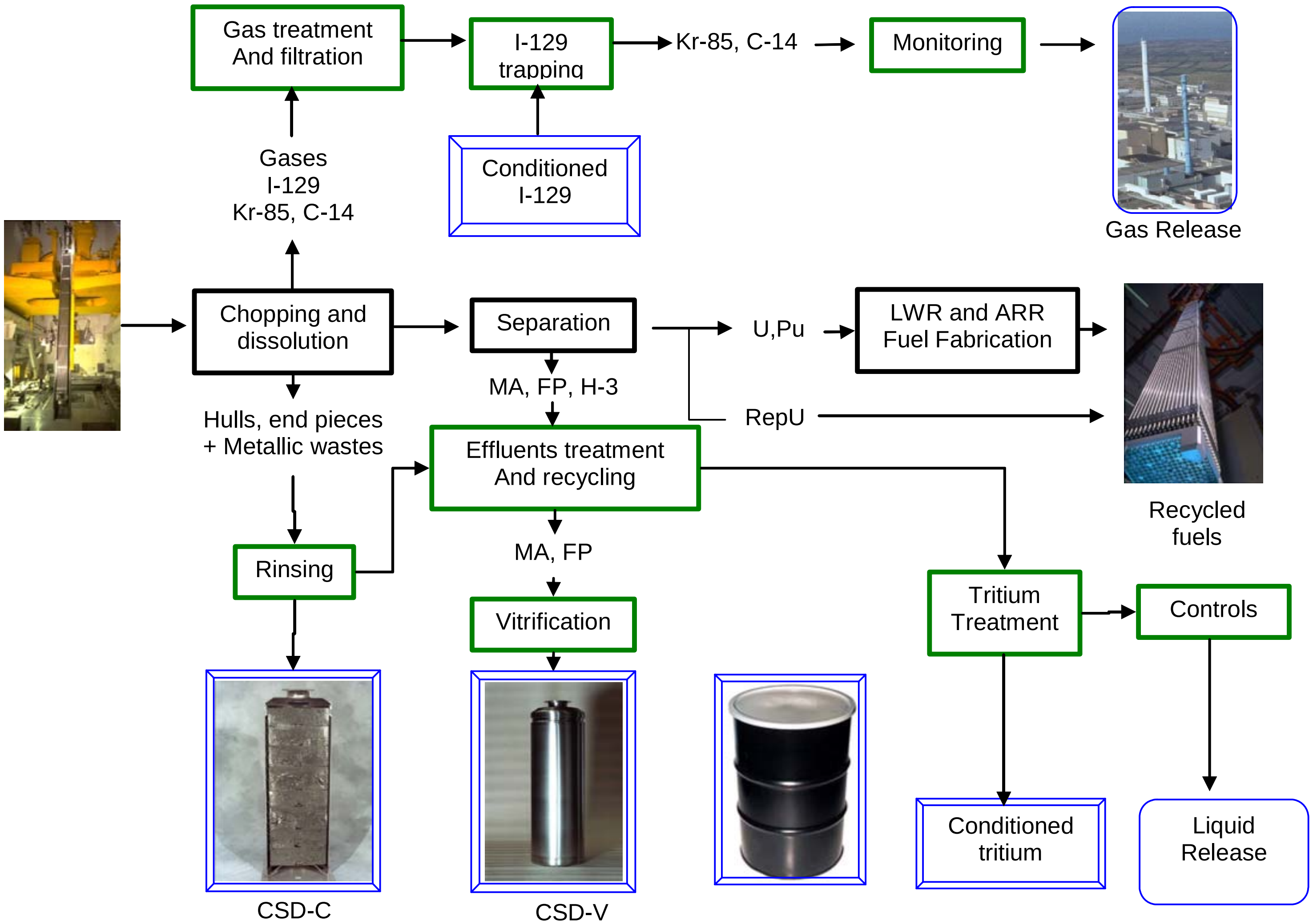

Figure 3 . General flowsheet for the initial CFTC recycling plant

DESCRIPTION OF THE PROPOSED WASTEFORM

Characteristics of the proposed glass product

Owing to the similarity in waste composition, the proposed wasteform would be very similar to the R7T7 vitrified wasteform currently produced at La Hague, and optimized for the expected effluent composition.

The liquid HLW produced by recycling UNF in the proposed design of CFTC will not vary widely in its radioactive composition since the waste is produced by a fixed single chemical separations process. This is in contrast to other types of HLW waste which are produced by any number of different chemical processes, such as the US Defence waste stored in tanks at Savannah River or Hanford. As a result, it is possible to formulate a single wasteform to accommodate the waste produced during decades of industrial operation of a recycling plant.

Furthermore, the separation process used in a recycling plant is optimized to minimize the amount of chemicals added to the HLW stream. The HLW stream is a concentrated nitric solution holding the fission products and actinides mostly as dissolved nitrate salts. The chemicals added to the process are selected so that they do not add large amounts of cations or undesirable anions to the HLW stream. The only cation added in a significant amount is sodium, but the effluent management in the plant is carefully optimized to decrease this amount of sodium to the minimum. Some traces of phosphates can also be observed, resulting from the degradation of the solvent, but these phosphates remain at a very low level, owing to the efficient solvent removal, regeneration and purification systems implemented in the plant. Only small amounts of iron, nickel and chrome are added as a result of the corrosion of stainless steel equipment in the plant. Finally, the effluent is vitrified in its acidic form without any neutralization, thus avoiding the addition of large amounts of caustic and the subsequent separate treatment of sludge and supernate. As a result, the waste loading for this type of wasteform is not limited by any inert chemical. It is limited essentially by the radioactive waste content.

The R7T7 glass formulation was adapted specifically for this type of commercial LWR fission product solutions (although for fuels with a lower burnup), with the noble metal –rich clarification fines. The baseline (reference) glass composition consists of 45.2 % SiO_2, 13.9 B_2O_3, 4.9 % Al_2O_3, 9.8 % Na_2O, 2.0 % Li_2O, 2.5 % ZnO, 4.0 % CaO, 2.9 % Fe_2O_3, 0.4 % NiO, 0.5 % Cr_2O_3, 13.7 % radioactive waste oxides (fission products, actinides, noble metals and Zr fines). The actual glass composition varies within allowed ranges around this reference composition. This R7T7 composition was designed to hold, at the maximum, 18.5 wt% radioactive waste oxides, or, equivalently, an overall waste loading of 28 wt%. This limit has been set to preserve a low crystalline yield (a few %) upon cooling.

The glass product is highly radioactive (predominantly ^{137}Cs, ^{90}Sr) with significant amounts of noble metals (up to 3 wt%, including strong beta-gamma emitters). The maximum $\beta\gamma$ activity at the time of production at La Hague is 28,150 TBq (760,000 Ci) per canister. The maximum contact dose rate can be greater than 10^5 rad/h. The maximum allowed heat output at the time of retrieval from storage is 2 kW per canister.

As a result of this very high activity content and heat output, it is necessary to limit the volume and diameter of the canisters in order to allow efficient cooling during storage and transportation. The resulting physical characteristics for these canisters are compared to the Defence wasteforms currently qualified or in the course of qualification in the US in Table I below:

Table I: Characteristics of the proposed commercial HLW canister compared to the US the Defense HLW

Characteristics	R7T7 wasteform (CSD-V)	US Defense HLW [3]
Canister height	~1.34 m	3.0 or 4.5 m
Canister outer diameter	0.43 m	0.61 m
Filled canister weight	< 500 kg	< 4,200 kg
Weight of glass in canister	~ 400 kg	up to 2000 kg
Maximum heat output at the time of shipment	2 kW	1.5 kW
Dose rate	can be > 10^5 rad/h	
Canister material	Stainless steel	Stainless steel
Closure method	Welding	Welding
Handling features	Concentric neck and flange allow the use of grapples	Concentric neck and flange allow the use of grapples

Several cask designs have been licensed to transport up to 28 CSD-V canisters nationally and internationally by road, rail, or sea, with acceptable heat-loads of 40 to 56 kW (TN28, TN81, TN85, Castor HAW...). [4]

The specification describing the La Hague-produced wasteform has been accepted by the French, German, Belgian, Japanese, Dutch and Swiss customers and their respective Competent Authorities.

Vitrification process

The CFTC vitrification process will draw heavily upon the very large design and operating experience accumulated in France in vitrifying exactly the same kind of effluents, as described in the literature [5], [6], [7] [8]. At the end of December 2007, the two R7 and T7 facilities (commissioned respectively in 1989 and 1992) had produced a combined total of more than 13,200 CSD-V canisters, of which more than 4,100 had been sent back to their foreign country of origin.

The French approach for the vitrification technology is to rely on small, modular equipment, that can be maintained using remote manipulators and cranes, and which allow optimizing the volume of corresponding waste. The facility design also allows performing major upgrades to the facility when needed [5]. Mechanical stirring of the melt has been implemented to process higher levels of noble metals: the current La Hague products can hold up to 3 wt% noble metals. The process proposed for the CFTC facility will use the latest French development in terms of melter: the process will use Cold Crucible Melters, one of which is in the course of implementation in one of the existing vitrification lines at La Hague to replace a hot wall induction melter.

Wasteform design and qualification.

The fission product solution to be vitrified will have characteristics very similar to the fission product solutions vitrified in France. So, the matrix proposed for the CFTC product will be very similar to the R7T7 glass composition, for which a full development and qualification program has been performed for several years at the Commissariat à l'Energie Atomique (CEA) in France. A large part of the experience gained during this program and many results will be directly applicable to the CFTC wasteform.

<u>Definition of the reference composition</u>. The formulation philosophy emphasized both processability (viscosity, melt temperature, general behaviour of the feed in the process) and durability (chemical durability, mechanical properties, and, most important in this case: resistance to irradiation and thermal stability). [9]. At first, in view of the feed composition and of its possible variations, a preliminary glass composition domain was defined. During this screening phase, the studies focussed on flexibility towards

expected composition variations, processability in the given technology, crystallization characteristics and leachability. The outcome of this screening phase was the definition of the baseline (or reference) composition.

Characterization of the reference composition. Once completely defined, the reference composition was characterized in more details, to better evaluate its processability and obtain data for the future product specification [10]. This characterization work was performed on samples prepared at bench-scale both with simulated and real waste, and also on samples taken from the pour stream and the canisters during full-scale inactive pilot operation, in order to demonstrate the applicability of the results obtained with inactive bench-scale samples to radioactive and industrial products. The effect of irradiation on the matrix was studied both by applying external irradiation and by doping some samples of inactive glass with short-lived actinides (^{238}Pu, ^{242}Cm and ^{244}Cm).

Sensitivity studies. In addition to the full characterization of the reference composition, sensitivity studies were performed to evaluate the allowable ranges around this composition [11]. Variations of individual and/or groups of components in the feed and variations in waste loading were tested, specifically targeting extreme compositions. The investigated effects dealt with processability (viscosity), chemical durability (by leaching) and thermal stability (lower crystallization temperature, maximum crystal yield). In parallel with this sensitivity study, tests on the inactive full-scale prototype platform were conducted to determine the sensitivity of the product to variations in processing parameters, such as melting temperature for instance. [12]

Effect of noble metals. The effect of up to 5 wt% noble metals on this composition was also studied [13]. As expected, this amount of noble metals led to a strongly non-Newtonian behaviour of the molten glass. The noble metals contents did not affect the crystallization characteristics (same phases found). The forward rate of dissolution determined by Soxhlet was not affected.

Acceptable composition domain. As a result of these successive development phases, an acceptability domain was defined that allowed realistic variations in the feed composition, and realistic performance for the various industrial tools, while maintaining the required level of glass quality and processability. Owing to the quite narrow expected composition range for the HLW effluent it was convenient to specify an acceptable composition domain within which the glass composition must fall, instead of establishing product-composition models for PCT response and liquidus similar to what is used in the US Defense facilities

Recently, studies have been started to optimize the formulation for fuels with higher burnups. The current reference fuel for La Hague facilities has a burnup of 33 GWd/MTHM. The waste loading in the glass is such that between 0.54 and 0.7 glass canister is produced for each ton of fuel. In the future, fuels with higher burnups will be reprocessed: 45 or even 60 GWd/MTHM. These fuels will contain higher amounts of fission products and minor actinides, a fact that could lead to the production of a larger number of canisters per ton of fuel. AREVA is now considering the possibility of increasing the waste loading for these solutions, in order to minimize the waste volume as much as possible. The results of this study will be directly usable for the proposed CFTC facility. The first step of this study has been to study the effect of increased self-irradiation on the long-term stability of the matrix, by fabricating glass samples doped with Cm-244 oxides at several levels (up to 3 wt%), in order to determine the effect of dose rate and cumulated dose on the material [9].

Other elements of the qualification process.

In order to ensure safety during the later life of the canister and compatibility with the downstream facilities, (handling, storage, transportation) several other topics had to be qualified which pertain to the canister and the canistered wasteform. Although the criteria were not expressed in the same way as in the US WASRD, and although the characteristics of the residues are different from those of the US Defence HLW, most of the same topics have been dealt with using comparable approaches.

The canisters are made of an austenitic stainless steel which has been specifically adapted for this application. A corresponding material specification has been designed for the base metal and for the filler metal used in welding operations. The fabrication procedure for the canisters has also been fully qualified and implemented by the agreed providers. The canister impact strength was evaluated according to the expected worst handling incident that could occur in the La Hague facility: a canister filled with 400 kg of glass was dropped 17 m onto the shock absorber at the bottom of a storage tube, or onto another canister already resting on the shock absorber. Even under least favourable conditions, container integrity was ensured, the containment remained intact, and recovery of the damaged containers from the well was possible.

Sealing. After filling, the cooled canisters are sealed using plasma arc welding. An automatic welding machine has been designed for this purpose, the procedure has been fully qualified through thorough testing, and specified allowable ranges have been defined for all the important process parameters.

Canister decontamination. The canisters are decontaminated using shot-blasting with an air-pressurized abrasive aqueous suspension of alumina and silica. During the development of this process, inactive and active testing was performed to show that this process did not affect the canister material.

Checking for removable contamination. The automatic contamination monitoring machine performs smear tests over the whole surface of the canister (in the US only 200 cm^2 are required). The operating parameters for the machine, the nature of the wipe test required, and the transfer coefficient from the container surface to the wipe were determined during qualification testing.

Quality Control

In addition to product development and qualification, it was necessary to design adequate quality control/quality assurance systems for the industrial production phase, to ensure:

- reproducible and predictable product quality and performance,
- reliable declared characteristics,
- traceability

Product quality[14] was defined with reference to:

- matrix quality.
- canister and canister closure
- removable contamination
- cooling and storage conditions

Matrix quality. In view of the level of radioactivity of the material, routine analysis and testing of the glass product would be virtually impossible: it would be a time-consuming operation, which would subject the personnel to un-necessary radiological exposure. Moreover, rework of the product, if non-compliant, would not be practical. As a result, it has been decided to rely on “before the fact” process control, in order to avoid conditions leading to an eventual non-conforming product. This approach, which is similar to the approach outlined for instance in the DWPF Waste Compliance Plan [15], requires extensive qualification work prior to facility commissioning, and the implementation of a rigorous quality control scheme.

Similarly to what is considered in the US, the major parameters that ensure matrix quality have been found to be composition and homogeneity. Failure modes and effects analyses were performed to identify fault conditions in the process that would impact glass quality as described above [12]. All the characteristic process parameters that could lead to these conditions were identified and systematic procedures, alarm thresholds, interlocks, and double-checks were implemented to decrease the risk of their occurrence, and detect them in time to allow implementing corrective actions. In the event of any discrepancy in the quality-related parameters, the process is switched to stand-by status, without feed or frit feeding, in order to implement the corrective actions. A specific analysis has also been performed to ensure that the detection time for each faulty condition was low enough to have only a negligible impact on glass quality. A two-year test program was implemented on a full scale pilot representing exactly the heart of the future industrial process, to evaluate the response of the system to upset conditions that could impact glass quality.

Glass composition is controlled by controlling several process parameters:

- HLW solution composition: the adjusted solution in the feed makeup tank is sampled and analyzed and calculations are performed to determine the target waste loading and heat release. The batch is cleared for transfer to the feed tank only if the adjusted batch allows producing an acceptable glass.
- Feed homogeneity: the feed tank is continuously stirred at a specified rotation speed in order to keep a homogeneous composition throughout the processing of the batch.
- HLW feed rate: the HLW solution is continuously fed to the calciner by a measuring wheel. Direct parameters as well as double-check parameters are monitored to ensure an adequate feed rate. The calciner parameters are also monitored (in the La Hague process), to ensure adequate calcine quality.
- Since, with this process, frit and solution are fed separately, the frit feed rate is also controlled and monitored in conjunction with the HLW solution feed rate, and verified by regular weighing of the feeding hopper. The frit is subjected to a strict procurement and acceptance process.

Glass homogeneity is achieved if:

- the melt temperature at the time of pouring is above a given threshold. This is monitored through several internal thermocouples.
- the melt homogenizing devices are operated properly
- for the specific situation at La Hague, one indicator of melt homogeneity is melt viscosity, which is assessed by monitoring the pouring on the basis of the weight increase in the canister.

In order to completely validate this approach for ensuring matrix quality as well as computing the matrix composition in the context of industrial operation, the Authorities required that, for each of the two facilities R7 and T7, a pour stream sample be taken during full active operation, and its characteristics be compared to the declared characteristics and to the specification. This was performed on 1992 for the R7 facility. The resulting specimens were shared for examination and analyses between two independent laboratories operated by CEA in France and JAERI in Japan. The agreement between the declared and measured composition was good and the crystallization characteristics of the glass were very similar to those observed on inactive R7T7 glass. [16]. The same operation was performed at the T7 facility in 1994, with similarly good results. The crystallization behaviour, static (MCC-1-type) and soxhlet leach behaviours of the active glass were very similar to those of the inactive simulant [17].

<u>Canisters and canister closure</u>. The quality of the canisters is verified at the time of procurement on the basis of the documentation, which include the compositional data of the raw material and of the filler metal, as well as the description of the fabrication operation. Visual and dimensional inspection is also performed upon receipt. During canister closure, the welding parameters are continuously recorded to

control their conformance to specifications for voltage, current, cycles, plasma gas flow rate and plasma torch rotation speed. If the parameters exceed the specified tolerances, a fully qualified repair procedure can be applied. The welding torches are procured according to the technical specifications and accepted only after test bench.

Canister contamination. The performance of the smear test machine is monitored. The wipes are procured according to specifications for fabrication, testing, packaging, shipment and assembly.

Canister cooling in the storage vaults must be performed so that the glass centreline temperature never exceeds 510°C. The canisters are stored in cooled wells. The air outlet temperature of the storage facility is monitored continuously and extra cooling systems can be activated if required.

Canister tracking: the uniquely identified canisters are tracked through the process using a video monitoring system and the operating control system.

Quality Assurance

A Product Specification document describing the expected characteristics of the product has been established. In addition to a description of the product, the specification provides some “guaranteed parameters” related to the above process functions which must be complied with in order to certify each canister as compliant. These guaranteed parameters include the chemical composition of the glass, the per canister radioactive concentrations for Cs-137, Sr-90, the per-canister actinide concentrations and some characteristics of the canister (dimensions, materials of construction, removable surface contamination, and heat release at the time of dispatching). [18]. The Glass Residue Specification has been approved in July 1986 by the French Nuclear regulatory Authority and by ANDRA, the organization in charge of HLW disposal in France. The specification has been subsequently approved by the customers in Belgium, Germany, Japan, the Netherlands and Switzerland.

A full Quality Control/ Quality Assurance program, complying with ISO-9002 requirements, has been implemented to ensure compliance and traceability for all important aspects relating to product quality. This program covers all aspects:

- procurements (glass frit, materials for the canisters, fabrication of the canisters, swab pads for smear test machine, welding torches)
- procedures (operations, maintenance analytical laboratory...)
- documentation: each canister is accompanied by complete QA documentation containing all the pertinent data relating to its production, including analytical results on the adjusted feed solution, the glass composition calculation sheet, and a description of processing operations for the corresponding glass batches. This documentation is thoroughly reviewed by AREVA before granting a certification for shipment and disposal.

The QA/QC program has been reviewed by ANDRA (the French National Agency for Radioactive Waste Management), acting on behalf of the Safety Authorities, during the Licensing Process for the facilities. An independent QA auditing company (Bureau Veritas), acting on behalf of AREVA’s customers, regularly verifies the quality of the final product through inspections and audits. The foreign customer also performs inspections at the time of dispatching. A very similar acceptance process seems to be in the course of implementation in Germany, for the VEK facility which is based on a LFCM technology. [19]

Leaching performance and behavior in disposal conditions

As seen above, per the French acceptance criteria, there is no explicit leaching requirement such as the PCT-A requirement for the US Defense HLW. Instead, there is a requirement that the Product Specification (which details an acceptable composition domain, and an acceptable operating range) be complied with. In effect, the glass compositions which belong to the acceptable domain are able to comply very easily with the PCT-A criterion. The R7T7 composition is a very robust composition which has actually become an international reference for commercial HLW immobilization.

As part of the acceptance process by the Safety Authorities, AREVA and the CEA have been required by the Safety Authorities to investigate the long-term behaviour of the glass in view of disposal in a geological repository. These studies have been going on for 15 years now, and an extensive data base has been accumulated for a large range of alteration conditions, from pure water to various repository-relevant conditions, from small samples to full size glass blocks [20-34]. The use of recent material characterization techniques has allowed considerable progress in understanding the glass alteration mechanisms at a very small scale.

For this well balanced glass composition in pure water, it is found that, after the initial stages of inter-diffusion and network hydrolysis, the dissolution rate quickly becomes controlled by the formation and consolidation of an alteration layer, whose porosity (and its progressive closure) is the major influent parameter. The alteration rates reached at this stage can become very low. This layer is formed by the progressive reorganization and re-condensation of the network after removal of the mobile and soluble species. This layer rapidly becomes an efficient diffusion barrier that slows the intrusion of water as well as the diffusion of leached species towards the bulk solution. At very long times, alteration is sustained only by the very slow diffusion of water through this layer towards the pristine glass, the formation of some phyllosilicate phases which slowly consume the silica from the layer, and the slow dissolution of the gel layer at the solution interface. For this glass composition, this layer, when formed in silica saturated conditions, is very stable, both chemically and mechanically, and is not altered by intermittent drying episodes. As a result, in conditions where silica saturation can be reached locally (such as quasi-closed systems where the silica released from the glass is allowed to accumulate), the glass alteration rate becomes very low in the long term. Moreover, this alteration layer retains most of the non mobile radionuclides (such as actinides, lanthanides, and transition metals for instance) but also a fraction of some more mobile radionuclides, such as U, Np-237, Sr, Ba, and even part of the caesium.

In order to de-stabilize this layer, it would be necessary to impose pHs that are higher than the pH reached by natural equilibrium between the glass and water, or to increase the temperature at values higher than those expected in the repository. The effect of glass composition variations on the formation of this protective layer has been studied, as well as the effect of glass cracking during cooling on the effective surface area exposed to water. The behavior of this glass in numerous environments (groundwater, engineered barrier materials including canister corrosion products...) has also been investigated

This knowledge has been incorporated into a model in the framework of the first French repository performance assessment, and the lifetime of a glass block has been estimated to be several hundreds of thousands of years.

In a deep geological repository, waste isolation is achieved essentially by the geologic barrier sometimes combined with robust engineered barriers. However, especially in degraded scenarios, the radionuclide containment properties of the wasteform itself can become significant for demonstrating the safety of the site. For a repository which will hold only commercial HLW glass, the accumulated knowledge base described above could be used and further complemented, to design a concept and engineered barriers that will preserve the glass wasteform for instance, and to help designing a source term model that takes credit for the retention properties of the matrix.

SUMMARY OF THE EXISTING HLW MANAGEMENT AND ACCEPTANCE PROCESS IN THE US

In the US, there is no commercial recycling of UNF. Two large scale vitrification plants have operated and one is under construction. The WVDP operated from 1996 to 2001 to convert reprocessing waste to glass. In Aiken, SC, nuclear waste from the processing of fuel to recover weapons Pu is converted to glass at the DWPF, which began operations in 1996 and continues today. At Hanford, defense wastes that have been stored in underground tanks are scheduled to be separated into low activity and high-level waste fractions, both of which will be converted to glass at the WTP. This facility is the largest of the US facilities and is currently under construction.

Glasses produced at these facilities are destined for the US geologic repository that is to be located at Yucca Mountain in the state of Nevada. To be accepted at the Yucca Mountain repository, the glasses have had to meet certain criteria as outlined at a high level in the waste acceptance specifications – requirements document (WAS-RD). The specifications are given in the waste acceptance performance specifications (WAPS). It is this latter document where the expected performance of the waste glass is specified; what tests need to be performed and what performance measures need to be met.

Testing programs from which data are generated to meet the WAPS have been long and entailed. For both WVDP and DWPF, these testing programs started in the early 1980s and continue to this day, although not at the same intensity as in early years of the program. These data demonstrate the ability to produce a consistent glass product with a uniform set of properties (compressive strength, leachability, etc), irrespective of the composition of the incoming waste. While not yet complete, the same kind of test program has started to support the vitrified waste product from WTP. The goal of these programs is to provide sufficient information to the repository that a total systems performance analysis can be made in support of a license application. This performance assessment must show that the release of radionuclides over the lifetime of the repository does not result in concentration in the near-by ground water that exceed license amounts.

A stochastic methodology is used as the basis for the performance assessment model for the Yucca Mountain site. In a stochastic model, a set of "look-up" tables are generated from a variety of sources, such as testing, modeling, expert opinion, and assumptions. These look-up tables are sampled while the model is calculating the release of radionuclides.

At the WTP, two waste glasses will be produced – low-activity waste glass and high-level waste glass. There will be approximately a factor of 10 more low-activity waste glass than high-level waste glass. The low-activity waste glass will be stored on the Hanford site at the Integrated Disposal Facility (IDF). This facility must retain radionuclides sufficiently well that ground water standards are met in the vicinity of the IDF. For glass, the radionuclide of interest is ^{99}Tc. Thus, the controlled release of ^{99}Tc from the low-activity waste glass must be sufficiently low as to maintain the ^{99}Tc concentration in the ground water just off the Hanford site below drinking water standards. To demonstrate this, a combination of glass testing, glass-water reaction modeling, and performance assessment is being performed. However, the approach to performance assessment is different at IDF than at Yucca Mountain. At IDF, a deterministic model has been developed. It is a fully coupled chemical transport code. Hence, the tests to support this performance assessment are significantly different than the ones that are used to support the stochastic model used at the Yucca Mountain repository. In the deterministic model, there are key parameters for which accurate values are needed. The more accurate and precise these values, the more accurate the performance assessment is. Therefore, the results form the tests that are performed in the IDF programs at Hanford are used to supply data for the model parameters.

DISCUSSION: INTEGRATION OF THE PROPOSED INRA WASTEFORM IN THE US CONTEXT

If the proposed recycling policy for UNF in the US is followed, the production of glass canisters from the 800 MTHM/yr CFTC facility may reach a steady rate of about 560 canisters per year (or 224 MT of glass per year), with characteristics that are quite different from those of the Defense HLW glass: the wasteform will emit intense radiation levels and generate a significant amount of heat. The radioactive contents will be several times that expected for the Defense HLW canisters. These different characteristics are the reason for some of the differences between the physical design of the proposed recycling HLW canisters and the Defense HLW canisters, on the basis of the considerable experience gained in Europe and Japan for this sort of wasteform. The proposed canister size, radioactive contents, irradiation characteristics, and heat output have proven to be safe and reliable. Fully licensed facilities are in operation to produce, handle, store [35], cool, obtain acceptance, and transport these canisters, with considerable and successful experience, which is directly relied upon by the INRA team to design the proposed wasteform.

As described above, the reference wasteform has been the object of a very detailed development and qualification program in France, and has been produced industrially for more than 15 years. This wasteform is well known and internationally recognized for its good performance. Although not performed in a US-based context, the qualification program has been very thorough, and based on the same principles as those considered in the US WASRD for ensuring a consistent and predictable glass quality: matrix quality relies essentially on the control of composition and melter conditions. In both types of facilities, it has been demonstrated that a priori quality control was feasible, provided that a stringent QA/QC program is implemented. One can then expect that the process and the matrix will have the best chances of achieving successful qualification in the US, while the benefits of the accumulated experience can be used to support the regulator and the disposal organization, and expedite the qualification work as efficiently as possible.

From the above discussion, it also becomes clear that the use of the GNEP approach changes the mix of waste in the Geological Repository from one that is UNF-dominated to one that is dominated by other waste types, essentially commercial HLW glass. This will then require an adapted approach for the repository. The corresponding concept and performance assessment will have to account for specific characteristics and radionuclide containment capability of the wasteform, in the specific geologic and engineered context. Additional testing to support adaptation and parameterization of the models will be needed, however the extensive characterization work performed in France for the “parent” wasteform will provide a sound and abundant basis to start this work.

CONCLUSIONS

One of the major aims of the GNEP and AFCI efforts is the overall control of the volume and environmental impact of waste generated from nuclear power, by avoiding the disposal of usable (and radiotoxic) material with the HLW waste. It has been emphasized by all the participants to the GNEP tender that the implementation of recycling would require some reorganization of the national regulatory and waste management schemes, and that wasteform qualification should be integrated into the early licensing activities for a recycling facility [36], [37], [38]. The proposed CFTC wasteform is based on a well known and recognized wasteform produced in France.

In this presentation we have demonstrated that, by combining the wasteform qualification experience gained for three facilities in the US (DWPF, WVDP, WTP) and the wasteform qualification and process knowledge developed in France (R7 and T7 at La Hague), the future CFTC HLW waste form should be easily compatible with the HLW management principles in the US. The above presentation has put into light some differences in the US and La Hague wasteforms and qualification processes, which result from the differences in the waste, the process and also the requirements for acceptance. However, the fundamental objectives are the same: ensure a reproducible, predictable quality for the wasteform throughout the lifetime of the plant, control its composition and activity content by a feed forward approach, carefully identify the processing parameters that enable good quality and control those parameters during production, implement a strict QA/QC program, ensure traceability, allow oversight by the customer, the disposal organization and the regulator.

Finally, the use of the GNEP approach changes the mix of waste in the Geological Repository from one that is UNF-dominated to one that is dominated by other waste types, essentially commercial HLW glass. This causes a change in the basis of the repository. The wasteform acceptance requirements will also have to be re-designed in accordance with this new situation. The extensive knowledge base and experience accumulated on the wasteform will be useful to support this overhaul, in order to make the best use of the optimization potential brought about by the concept of recycling (less actinides, smaller volume, waste loading, different heat output vs time, interesting long-term behavior...). This optimization will need to be performed in close cooperation between the disposal organization, the regulator, the wasteform producer, and the wasteform owner, as it has been demonstrated during the qualification of HLW wasteforms both in the US and in France.

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