Management of HL Deposits for the D&D Of UP1 Reprocessing Plant - Retrieval and Treatment Scenarios - 15295

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ABSTRACT

Built in 1958 on the Marcoule CEA site, the UP1 facility, was the 1st French industrial (500 t U / year) reprocessing plant based on the PUREX process. The plant, which has stopped operation in 1997, constitutes with its auxiliary workshop (vitrification workshop, glass canister storage), one of the largest and most challenging decommissioning project in France.
In about 40 years of operation, various type of fuels elements such as metallic uranium, U-Al fuel rods, Phénix breeder elements or PWR fuel have been reprocessed, thereby requiring several plant and process adaptations. Resulting from these complex process chemistry and operation history, significant fouling of the chemical plant circuits and components has occurred. The fission product (FP) concentration process is particularly concerned. Preliminary investigation performed in 2010 in the evaporator's feeding tanks revealed the presence of significant amounts of sludge and dose rates as high as 250 Gy/h. Therefore a thorough investigation campaign, including visual inspection, volume quantification, deposits sampling, and radiological characterization of the components of this workshop has been initiated in 2013. Chemical analysis performed so far show that the solids are mainly composed of zirconium phosphates, "MoZr" compounds and nitrate salt precipitates. But the presence of insoluble fines arising from the dissolution workshop and organic residues resulting from the TBP decomposition has also been evidenced in some of the tanks.

The numerous constraints associated with the management of such highly active sludge, with Cs-137 specific activity ranging from 1 to 10 GBq/g, and a complex physico-chemical composition, lead CEA to investigate the implementation of an in situ conditioning treatment able to produce a waste package complying with existing evacuation routes and/or on-site storage facilities. The full paper presents an overview of the UP1 D&D project and then focuses on the possible dismantling scenarios for the FP concentration workshop, including the evaluation of different strategies for the highly active deposits management. The investigations and deposits characterization program will be developed so as the sampling methodologies that has been specifically tuned. In the light of the best actual knowledge of the deposits properties, the recovery and conditioning treatment scenarios will be exposed, considering, as a case study, the development of a specific vitrification tool to be implemented *In Situ* and specifically designed to match the requirements and constraints of the D&D project : adequately dimensioned, compact, flexible with respect to the feed composition, for a limited duration of use and designed to be dismantled.

INTRODUCTION

In the 40 years of operation of the UP1 plant, various type of fuels elements such as metallic uranium, U-Al fuel rods, PHENIX breeder elements or PWR fuel have been reprocessed, thereby requiring several plant and process adaptations. In 1964, a new dissolution workshop, working in continuous mode, the so-called MAR200 facility, has been put into operation in order to increase the overall plant treatment capacity. Resulting from this complex process chemistry and operation history, significant fouling of the chemical plant circuits has occurred and insoluble particles or precipitates have settle down and accumulated in different vessels and components of the process. Since the shutdown of UP1 production in 1997, significant D&D progress has been achieved, especially in the medium active area where concentration, purification and conversion of U and Pu took place. However in the high active area, the FP concentration workshop, also referenced as workshop 71, has been kept into operation until 2009 in order to treat liquid effluent arising from the specific rinsing program of the nearby vitrification and FP storage facility (SPF-AVM). Only very limited flushing operations were performed on the main circuit and components of the UP1 plant prior to the definitive shutdown at the end of 2009. Following these operations, significant increases of dose rate were observed on some of the evaporator's feeding tanks in the FP concentration workshop.

CHARACTERIZATION OF HL SLUDGES AND RESIDUES

Nature and Origins of the Highly Actives Deposits in the FP Concentration Process

The purpose of the workshop 71 was to concentrate, by evaporation, the acidic FP solution arising from the U/Pu extraction process prior to their conditioning in a glass matrix at the Marcoule vitrification workshop (SPF-AVM). This workshop was put into operation in 1958. It was initially composed of 3 production lines (A, B & C), each of them including a 10 m^3 receiving and feeding tank (referenced 71.21) equipped with cooling pipes (see Figure 1), a 400L buffer tank, 2 evaporators (respectively referenced 71.26 and 71.46) and their condensers. A simplified diagram of the process is presented on Figure 2. The production line A was definitely stopped in 1965, decontaminated and subsequently dismantled in the early 90's while the evaporator 71.46B was modified and converted to the recovery and concentration of nitric acid. At the present time, 3 evaporators remain in the workshop: 71.46C, 71.26B and C, which respective useful volumes are 18, 15 and 11 m^3.

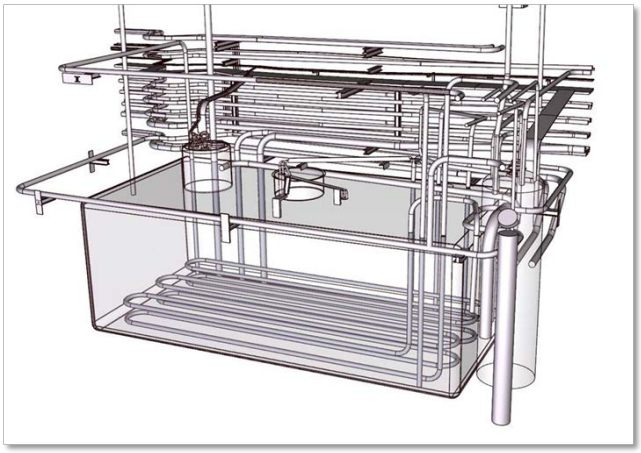


Figure 1 : Receiving and feeding tank 71.21A,B,C

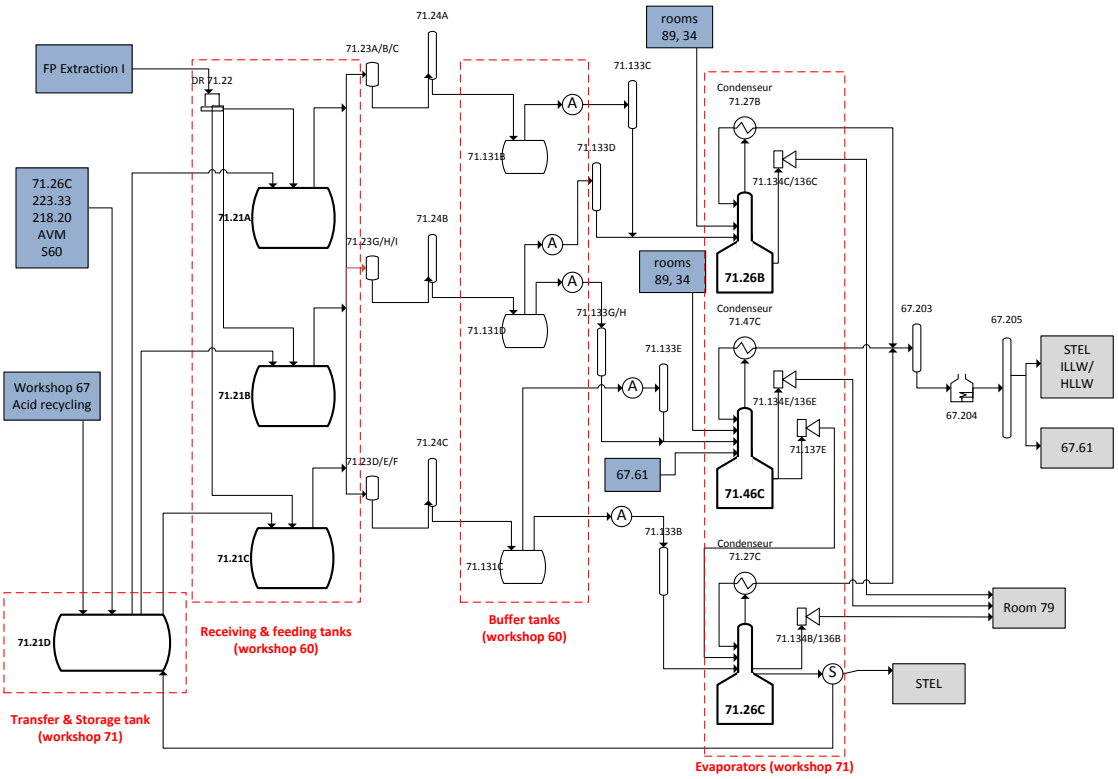


Figure 2 : simplified diagram of UP1's FP concentration workshop

Preliminary investigations were performed following the definitive workshop shutdown. These investigations mainly used available instrumentation and access(es). For instance, the volumes of deposits remaining in tanks 71.21A,B,C have been estimated by the standard addition method using the existing bubbling level sensor, while dose rate profile along the tank were measured through an instrumentation passage hose. These results allowed estimating roughly the volume of deposits remaining in the equipment and to calculate the associated activity using the MERCURE code. However these investigations were far too restricted and constrained to obtain sufficient and reliable data. In order to consolidate these preliminary data, a more comprehensive program of investigation, including a sampling of the sludge/solids, was launched in 2010 in the tanks 71.21 A/B/C/D. This program, which will be applied to all process vessels and equipment of the workshop, aim to :

- Determine the physical and radiological status of the different process components,
- Evaluate the volume of deposits remaining in each equipment,
- Determine the radio-chemical composition of the deposit,
- Determine the physical and mechanical properties of the deposits.

Those investigations are essential to define the deposit's retrieval processes, identify constraints associated with their interim storage, launch the development of a suitable conditioning matrix, and subsequently define a detailed scenario for the dismantling of this workshop. When defining investigation scope and methodology, the following constraints have to be taken into account:

- Vessels and equipment accessibility,
- Ambient dose rate and associated personal exposure risks,
- Radiation compliance of visual inspection equipment ,
- Samples conditioning and transport.

To perform these investigations, 2 specific accesses have been created for each tank by core drilling through the shielding slabs and then through the vessels (see Figure 3).

Dose Rate Profiles in the Casemate and Tanks

Based on the first activity estimates (100 TBq per tank in 2004), the dose rate at the top of the casemates and in the tanks were evaluated in order to define the diameter of the core sampling and to develop sampling devices. The core sampling diameter should both allow to operate above the equipment with acceptable conditions and to introduce investigation's equipment. Two 200 mm diameter accesses have been created. The dose rate mapping was performed in two phases: in 2010 in the casemates and in 2011 inside the tanks after trepanation. Table I shows the obtained results.

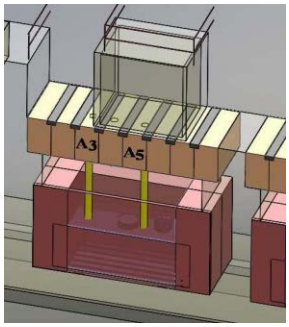


Figure 3 : Access into tanks 71.21 ABC

Table I: Measured dose rate in evaporator's feeding tank (campaign 2010)

	71.21A	71.21B	71.21C	71.21D
Ambient dose rate - Casemate top (Gy/h)	4	5	3	4
Dose rate in contact with the tank (Gy/h)	23	20	15	10
Dose rate in contact with the deposit (Gy/h)	**245**	**150**	**38**	**225**

Based on this new dose rate measurements, previous deposit's volume evaluation and calculation using the MERCURAD© code, the tank's deposit activities were subsequently re-estimated between 1000 and 1500 TBq per tank. This new input data was used for re-designing visualization equipment and re-sizing the sampling means and their associated biological protections.

Evaluation of The Deposit's Volume in the Tank

Televisual inspections aimed to estimate the deposit volume in the equipment, monitor the sampling operations and visually assess the sludge texture and mechanical behavior. Given the level of radiation in the tank, a radiation-tolerant camera was purchased. Its technical specifications are summarized in Table II.

Table II: ALLRAD video camera features.

	Diameter : 110 mm
	Optical zoom : x3
	Weight : 3.7 kg
	Panoramic view +/- 180°
	BW camera
	Radiation resistance: 1 kGy/h

The volumes of deposits are estimated by two methods:

- Construction plans were used for the tanks 71.21A and 71.21D, Figure 4 shows the deposits height with respect to the tank cooling pipes position,
- In tank 71.21B, a graduated rule was remotely introduced to measure the deposit height at different locations.

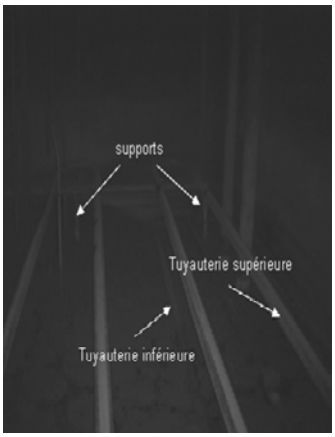


Figure 4 : Deposit's volume estimation (Tank 71.21A)

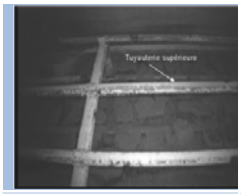
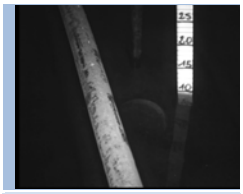
The resulting volume estimates, which are presented in Table III, have a high uncertainty of about 30%. Early return of experience on others CEA D&D projects has shown that photogrammetry is a very accurate (+- 10%) and cost-effective technology to quantify visible deposits, solid residues and heel in process vessel.

Unfortunately it can hardly be implemented in this project since:

- the deposits are maintained underwater for radioprotection reasons,
- accesses inside the vessels are very narrow,
- the camera has to be compliant with the high radiation field.

Televisual inspections also evidence significant differences in deposits mechanical and physical properties in the 4 tanks: a whitish crust in 71.21D, an overburden granular deposit in 71.21A, blackish and pasty slurries in 71.21B and C.

Table III: Estimated deposits volumes.

	71.21A	71.21B	71.21C	71.21D
			Not usable	
Volume (L)	500	400	< 1000	160

Sampling Equipment

Various techniques exist to collect material out of the equipment: vacuum or mechanical system. Vacuum systems are suitable for homogeneous fine-grained dry or liquid deposit type. In the case of a heterogeneous deposit, for which the physical properties and behavior are unknown, a mechanical system is recommended to avoid segregation phenomenon during sampling.

Figure 5 : mechanical sampler

Considering the high dose rate, the limited accesses available and deposits' nature, specific tools were developed in order to be able to sample materials in the tank. An air driven mechanical device with a useful volume of 10 cm3 was used. The mechanical sampler's volume was intentionally limited to minimize risks associated with personal exposure or non-compliance with internal transport regulations or laboratory activity limits.

The mechanical sampler has a transparent window allowing checking with the ALLRAD camera

that the sampling has been successful. The characteristics of the samples are summarized in Table IV. The developed sampling device is fully air-driven. Its deployed length is 7.2 meter, with a diameter of 60 mm and a weight of 38 kg. The maximum horizontal deployment length is 1m from the axis. A sleeve connect the tank opening with the top of the slab, thereby preventing contamination spreading outside the tank and allowing in situ decontamination of the long handling tool. Six samples were collected per tank. Figure 6 shows the samples locations.

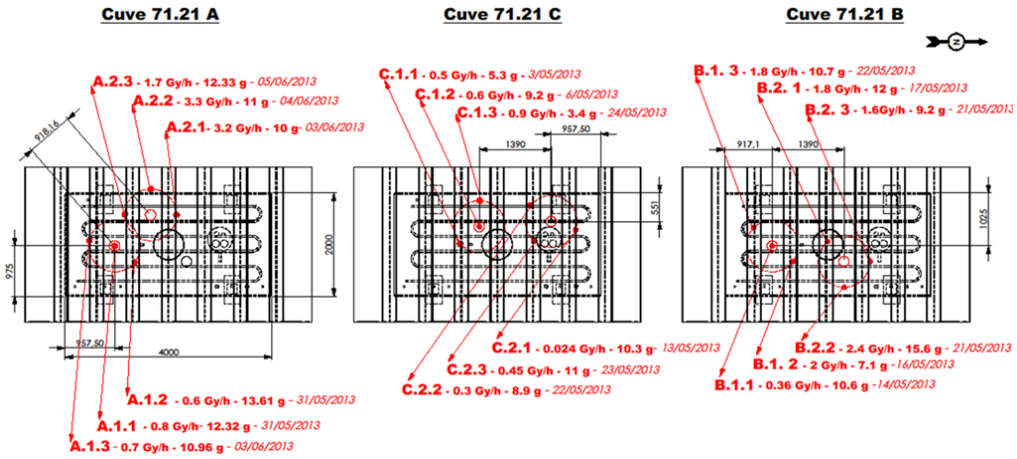
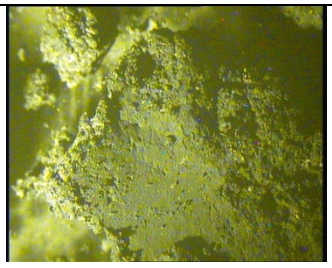
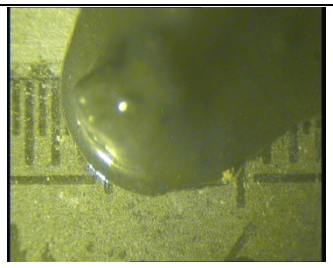


Figure 6 : Sampling plan for tanks 71.21A, B, C

The samples have been collected in the summer 2013 and are being analysed at the NUCLAB hot laboratory at the Marcoule site. The mechanical sampler are packaged in a water tight silicon case, placed in a CTX bin and transferred to the laboratory in PADIRAC shielded transport cask. A specific tool has been designed to open those cases with a remote manipulator arm. The sticky nature of the deposits sampled and the geometry of the sampler somehow impaired the recovery process. Table IV shows the main characteristics of the deposits right upon receipt and opening in the laboratory hot cell.

Table IV: Characteristics of the collected deposits

71.21A	**71.21B**	**71.21C**
10 g < sample weight < 15 g 1 Gy/h< dose rate < 3.5 Gy/h Aspect : Black mud	7g < sample weight < 15g 0.5 Gy/h < dose rate < 2 Gy/h Nature : Black solid	5g < sample weight < 11g 0.1 Gy/h < dose rate < 0.9 Gy/h Nature : Highly liquid black mud
		

When drying in the hot cell atmosphere, significant changes of samples colour and texture are

observed : they become whitish and highly powdery.

Laboratory Analysis

An extensive radiochemical and physical analysis program has been implemented including:

- Elementary analysis of the deposits composition,
- Isotopic composition and specific activities of α and βγ emitters,
- Speciation and quantification of solvent degradation product,
- Physical properties, such as bulk density and particle size distribution.

The main objectives of this program are :

- To strengthen the radiological inventory of workshop 71,
- Assess as accurately as possible, the fissile material concentration in the deposits,
- Gather data allowing to launch an R&D program on conditioning of these HL deposits,
- Assess the compatibility of the raw deposits with existing or future interim storage facilities,
- To assess mechanical, hydraulic and/or aeraulic behavior of the sludge.

Despite numerous technical difficulties, the radiological and chemical analysis of the 6 samples taken in each of the tank 71.21 A, B and C have been completed. The major issues which have been encountered are:

- Loss of material during the samples recovery and conditioning steps (in Hot-cell),
- Samples texture evolving quickly over time from a black sticky sludge to dry whitish aggregates,
- Highly disseminating samples requiring regular tools decontamination and hot-cell clean-up,
- High dilution factor required to comply with activity limit of analytical equipment in glovebox.

The main results of the radiological analysis are summarized in Table V.

Table V : Radiological characterization of HL deposits in tanks 71.21 A, B and C

Nuclides	Specific activity of the deposits (Bq/g of dry sample)		
	71.21A	71.21B	71.21C
Cs-137	$2.10^9 – 1{,}3.10^{10}$	$2.10^9 – 1{,}1.10^{10}$	$4{,}4.10^8 – 1{,}1.10^{10}$
Sr + Y	$2{,}3.10^6 – 1{,}8.10^7$	$1{,}4.10^7 – 2{,}7.10^7$	$5{,}6.10^6 – 8{,}5.10^7$
Total α *Incl. Pu^{239} - Pu^{242}*	$4.10^6 – 1{,}2.10^7$ (42 – 57%)	$7{,}1.10^6 – 2{,}1.10^7$ (35 – 48%)	$1{,}2.10^6 – 2{,}7.10^7$ (45 – 55%)

In the 3 tanks, Cs-137 represents between 95% and 99% of the total activity of the deposits. Others γ nuclides detected includes Ru-106, Rh-106 and Sb-125 but their contribution never exceed 1% of the γ spectrum. Pu Isotopic composition combined with α spectroscopy measurements allow to quantify the Pu content in the deposits. Average Pu concentration are ranging from 0,8 mg/g (dry sample) in tank 71.21A to 1,7 mg/g in tanks 71.21B and C. A larger discrepancy in nuclides activities is observed between the samples of tank 71.21C in particular with respect to the Pu content (1,7 mg/g +- 1,2). It is also noticeable that the activity ratio Sr-90 over Cs-137 is extremely low.
The results of the chemical characterization of the deposits are synthetized in Table VI.

Table VI : major constituents of the HL deposits

Element	Elementary composition of the deposits (%w of dry sample)		
	71.21A	71.21B	71.21C
Zr	14 – 17	10 – 18	16 – 21
P	7 – 11	4 – 11	5 - 36
Si	3 - 10	4 – 23	3 - 33
Mo	3 - 7	0.4 – 8	3 - 30
Fe	0.5 - 3	1 – 2	1 - 4
Al	0.5 – 1.4	0.5 – 1	0.5 - 2

The main elements (about 50% in weight) composing the deposits are Zr, Si, Mo, Fe and P. Phosphorus exists as phosphate, which is the end-product of the solvent degradation. Comprehensive reviews of these results, of the reprocessing literature and of the operating history lead us to conclude that the deposits are mainly composed of 4 types of components:

- Zr complexes : $Zr(HPO4)_2$,nH_2O and/or $ZrMo_2O_7(OH)_2$
- Cs complex of the type $Cs_xPMo_yO_z$
- Pu complexes : Pu phosphate and/or Pu Molybdate,
- Oxides and hydroxides : ZrO_2,nH_2O, PuO_2,nH_2O, MoO_3,nH_20.

Except for $Zr(HPO4)_2$, which presence is confirmed (in tank 71.21A) by a constant Zr/P molar ratio close to 0.5, the respective amounts of the different complexes within the deposits still has to be assessed by solid state analysis. The $Zr(HPO_4)_2$ solids have a very high affinity with Cs-137 (zeolites), which could explain the activity segregation observed and the rather low Sr-90 activity measured in the solids. This property has to be considered when defining the sludge recovery process. Again, a larger discrepancy in the sludge composition is observed 71.21C.
Finally, due to the limited sampler volume, the resulting amount of dry material was sometimes unsufficient to achieve the complete analysis program. Therefore a complementary sampling campaign is being scheduled, to complete the physical characterisation of the HL deposits, to assess the presence and the proportions of the different type of complexes and to define a truly representative composition of the sludge.

WASTE MANAGEMENT STRATEGIES FOR THE D&D OF WORKSHOP 71

The main challenge of this D&D project is clearly the management of these HLW slurries. Considering on the one hand the extremely high specific activity of the deposits (10^9 to 10^{10} Bq/g) and the other hand the definitive shutdown (12/12/2012) of the Marcoule vitrification workshop, few options exists at the present time for interim storage and/or conditioning of these HL waste (see Table VII).
The heterogeneity of the deposits, the significant amount of TRU elements and the suspected presence of solvent degradation products raise additional issues with respect to the compliance with the interim storage waste acceptance criteria, such as : the radiolysis rate limit, waste package filling rates and data and product quality objectives.
Therefore CEA considers the retrieval operation shall include a “pre-treatment” stage aiming to minimize “non-compliance” risks.

Table VII : French radioactive waste classification and disposal routes

Waste type	Criteria		Disposal (ANDRA)	Interim storage (CEA facilities)
	α activity	βγ activity		
VLLW	Typically < 100 Bq/g IRAS Index < 1		Surface disposal (Morvilliers)	Producer
ILLW – SL	< 3,7 GBq/t (waste package)	< 330 GBq/t (waste package)	Surface disposal (Centre de l'Aube)	Producer
LLW – LL	< 3,7 GBq/t (waste package)		Dedicated near surface disposal (graphite, alpha-rich waste, Radium- bearing)	CEDRA, EIP, EIP-HB[a]
ILW – LL	>3,7 GBq/t (waste package)	Typically $10^6 – 10^9$ Bq/g	Deep geological disposal CIGEO > 2025	CEDRA, DIADEM[b], EIP, EIP-HB[a]
HLW - LL	α and LL β emitters heat production > 13 W		Deep geological disposal 2025 < HA glass < 2042 2065 < MA glass < 2070	DIADEM[b], SVM[c]

[a] bulk waste, available in 2026 (Marcoule)
[b] bulk waste (no glass canisters), available in 2019 (Marcoule)
[c] glass canisters storage facility (Marcoule site)

Another significant issue to consider in the scenario is the "stop" of the bituminization. The former Liquid Waste Treatment Plant in Marcoule (STEL) will be refurbished and co-precipitation sludge will be incorporated into cement from 2016 on (STEMA project). The plant capacity to treat HLLW is subsequently reduced and the use of liquid for pre-treatment, sluicing and/or retrieval of the slurries must be considered as a significant project risk.

Risk mitigation actions would include:

- launching a leaching test program on "real" samples to assess radionuclides solubilization and define appropriate working conditions,
- foreseen worksite effluent treatment system,
- support R&D effort on inorganic ion-exchange media (functionalized zeolite),
- Minimize effluent volume by recycling and/or using existing supernatant for sluicing operations.

Figure 7 : Tirade container

An additional major issue is the (on site) transportation the HL slurries. The feasibility of docking and manipulating, in the vicinity of the FP concentration workshop, B-type transport cask, such as Tirade (see Figure 7) or IR500 (see Figure 8), which respective weight without loading are 20t and 33t, still has to be demonstrated. At this early stage of the studies and considering that waste acceptance criteria of future interim storage facilities such as DIADEM are subject to change, project planning has to be "secured" by implementing a "buffer storage" in the facility itself.

Figure 8 : IR-500 (licensed for on-site transport of Phenix fuel)

Finally for the purpose of the scenario study, the following evacuation routes are considered:

- DIADEM,
- EIP – HB (bulk material),
- EIP extensions (blocked waste),

- SVM (glass canisters),

Based either on existing (SVM, EIP) or project (DIADEM, EIP extensions) waste acceptance criteria specification and state-of-the-art analysis of "qualified" conditioning matrix, the waste packages streams generated by the deposits retrieval operations have been estimated for each "available" route either as "powdered" or as conditioned waste. As shown on Figure 9, the cementation of the HL deposits would certainly lead to a severe increase of the waste volume and subsequently higher management cost.

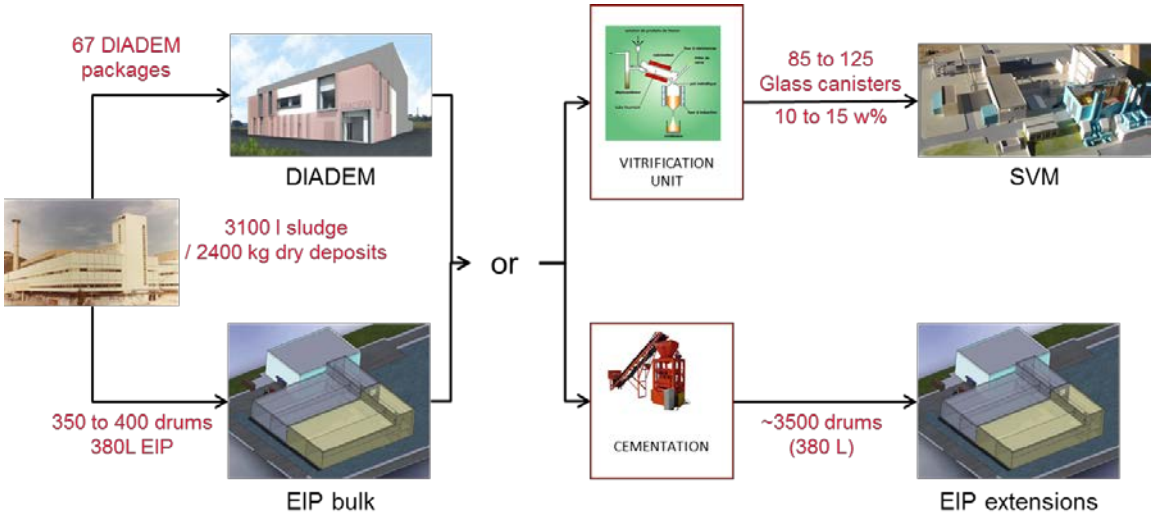


Figure 9 : Waste packages streams generated by the recovery of the HL deposits

To define the most cost-effective disposal route for this HL deposits, one has also to consider the interim storage durations for both the raw waste and the conditioned waste. Indeed, according to the French national plan for radioactive waste management, C type waste will be much earlier disposed of in the geological repository than B type one (see Table VII). Therefore when considering the vitrification of the HL residues, constraints associated with the production of HL glass should be carefully evaluated with respect to the additional decades of interim storage as IL glass canisters.

Finally, in order to secure the project and define a "baseline" scenario, CEA has developed a comprehensive detailed study and engineering program supported by R&D actions in different CEA labs.

The program priorities have been defined as follows:

- Investigation and characterization of the residues in the evaporators,
- Realization of an additional sampling campaign in evaporator's feeding tanks for completion of the characterization program,
- Realization of preliminary test on active samples to support the definition and the development of the HL sludge retrieval process(es),
- Assess the feasibility of the implementation of heavy infrastructures (hot cells, docking of B-type transport cask …) in the vicinity of workshop 71, through a detailed study of UP1 civil engineering works,
- Study the feasibility for the transport, treatment and conditioning of HLLW at La Hague,
- Study the feasibility of the HL deposits conditioning using a "dismantling" vitrification tool.

DEVELOPMENT OF A DISMANTLING VITRIFICATION TOOL – THE "IN CAN" PROCESS

The waste streams from dismantling operations described above are for some of them today identified as a potential issue in terms of handling, transportation, adapted conditioning matrix or evacuation outlet within the existing regulations. These particular conditions would justify the evaluation of an in situ treatment capable to produce a waste package complying with existing evacuation outlet.

The chemical composition of these deposits is not fully known at this time (due to sampling and analysis challenges) ; but it can be seen that they are mostly composed of nitrates and minerals : fission products with cesium as principal activity contributor, actinides, Mo, Zr, corrosion products, nobles metals (Ru, Pd), metallic fractions, organics traces. Cement matrixes may be excluded due to the high level of activity (several hundreds of Gy/h at contact producing hydrogen release by radiolysis, non-compliant with final repository) and huge waste volume expansion. In this context, a glass matrix seems able to bring a satisfying answer as the glass material and the vitrification processes have demonstrated to be adapted to high level waste and can be flexible enough to accommodate a variable waste stream.

This type of tool has not been developed today, that is to say: for high level waste, designed for a short duration of use, as a decommissioning tool, able to be installed and operate inside a decommissioned cell, aimed to be dismantled just after the treatment operation and producing a small amount of secondary waste. CEA started to study the development of such tool.

Discussion on Feasibility

The current analysis available of the chemical elements (cations and anions S, F, Cl) is a priori sufficient to start considering the feasibility and process design orientation. The presence of corrosive species (sulfur, chlorine, molybdenum, fluorine…) has to be taken into account for the corrosion aspects and gases treatment. The presence of organics, if any, in large quantities must be known in order to: manage their oxidation prior to incorporation into the glass matrix of the mineral part, adapt the redox of the glass and the gases treatment design.

The main point about the search for a glass composition suitable for the application will be its flexibility considering the waste composition variability. Indeed, as it is likely that the composition of the waste will not be fully known, the purpose of the glass design will be to obtain a matrix which is able to guarantee a description with a wide variation in the composition as input. As the amount of waste to treat is low, a high waste loading should not be a main objective. From a summary analysis of the types of waste, a waste loading of about 10-15% by weight of the waste form of oxides should be worth reasonably achievable

The amounts of waste to be treated are low, of the order of a few hundreds of kilogram; the rate of treatment could therefore be compatible with an installation of small size with low capacity. The use of a small scale facility would, in particular:

- Facilitate the implementation, consistent with an introduction into an existing dismantling site in the form of a "vitrification skid",
- Develop a design to "limited use", especially in choosing the materials and equipments just needed for a short operating time and easily dischargeable,
- Reduce security constraints by associating the equipment to a "decommissioning tool" and not a vitrification shop. This tool would be dismantled on site right after use,

- Consider the equipment for a given site, economical enough to have better equipment by site rather than moving waste between sites (which is likely infeasible in some cases),
- Reduce investment.

The process feeding may be performed in solid phase or in solution / suspension in water, as long as the intake flow rate into the furnace is controlled and that the recovery of the upstream waste in equipment is feasible. A batch process can also be considered to simplify the feeding process. Feeding has to be considered with the possibilities of recovering of the waste (possibility of dissolution, size of grains …)

The gases treatment is to study according to the details of the waste composition but is not seen as a stage gate. If an effluent treatment station exists, a liquid gases treatment producing compatible effluent is preferred as the wet OGTS is industrial and proven to achieve desired DF (decontamination factor) for volatile species. If no effluent treatment station exists, a wet OGTS is still interesting to be considered if the amount of waste to treat is low. In this case, considering a smart liquid recycling management from OGTS, only a very small amount of low activity effluent could be generated, compatible with an existing outlet. Dry treatment, however, can also be considered incorporating an internal recycling cycle in the vitrification process.

To take into account existing evacuation outlets, it should be considered that the glass containers must be packed in an existing format (For Marcoule, Standard French glass canister CSD-V or Marcoule Vitrification Shop [AVM] Canister). The practical limits are: too small sizes that would lead to too frequent handling, too large sizes that increase stress on the design of the system and lead to leave the melting pot for too long in the case of an in can melting process (risk of corrosion).

Technology Assessment

Existing high level waste melters can be classify in different categories depending on their heating mode (joule or Induction), their crucible design (ceramic, hot or cooled metallic walls) and their feeding mode (continuous or non, liquid or solid fed). These different designs have all been used in commercial operation and have their own advantages and drawbacks, the main points to consider are:

- Compactness
- Specific capacity
- Tolerance to particular glasses (corrosives, high temperature, …)
- Complexity (heating, draining)
- Implementation complexity
- Equipment Lifetime
- Flexibility of operation (Tolerance to stop & start, idling, …)
- Industrial experience
- Remote handling possibilities
- Secondary waste
- Cost

If we consider the main characteristics we are looking for the application to a decommissioning vitrification tool, our vision is that this type of process would be pretty close to what CEA has developed for an internal application: the IN CAN batch melting process [1]. In this process, the container is used as a crucible ("in can") that is renewed with each batch:

- As the crucible is renewed each batch, that greatly simplifies the equipment and makes it very robust :
    - There is indeed no need to drain the glass,
    - Corrosion problems are limited by the batch limited time,
    - The secondary wastes are limited to the heater as there do not remain after use any components that have been in contact with the molten glass,
- Thanks to its small size, the crucible can be easily heated by a simple and robust resistance heated furnace, inexpensive, easy to maintain, easy to implement. This heating mean allows to have a metallic fraction in the canister,
- The process has shown considerable flexibility as it will for example for the CEA application be exploited only with two shift teams, idling without operators during night shifts,
- The compactness and basic technology of the IN CAN melter makes it a low investment equipment compare to other technologies,
- The IN CAN process has also drawbacks :
    - It is capacity limited due to its small size and limited temperature ; this is not a disadvantage for the intended application,
    - High temperature glasses are not reachable. In the intended application it will be more important to have a low temperature glass to limit Cs volatility,
    - It has to be paid attention to extreme corrosive glass and glass composition / canister material have to be designed together,
    - In the simplest version of the IN CAN, the homogeneity of the glass is only given by thermal convection. If enhanced homogeneity is needed, extra devices or furnace design can be necessary.

The existing pilot is composed of a vitrification furnace, a particle separator, a condenser, a scrubbing column, and en exhaust blower, installed on three levels. The unit capacity is approximately a little bit more than one hundred kilograms of glass per week.

Figures 10 and 11 show the process diagram and a picture of the pilot. Glass is produced at about 1100°C in an expendable melting pot which becomes the disposal canister at the end of the test. The melter is supplied directly with the liquid vitrification feed solution and with glass frit. The feed streams are supplied through a connecting sleeve between the melting pot in the furnace and the particle separator. The furnace is designed and dimensioned for use in a glove box. The dome comprises nine 5.5 kW heating elements secured to the top of the furnace and extending vertically around the melting pot. The furnace has only one heating zone. It is also equipped with a thermal expansion absorber to prevent deformation of the melting pot. For initial application, the pot is

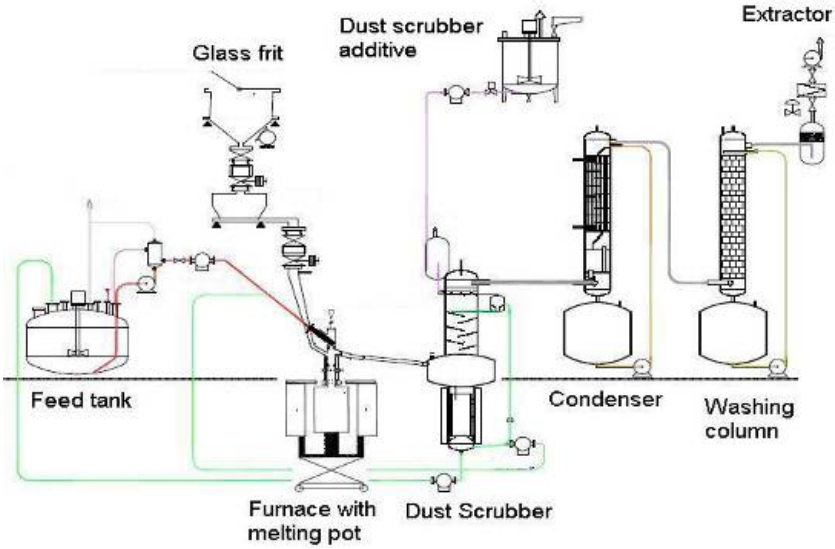


Figure 10 : Diagram of full-scale pilot in direct vitrification configuration

filled with 108 kg of glass. Physically the entire volume of the pot is about 50 L. Two pots can be stacked in a standard CSD-V canister. The entire process is depressurized. Particulates in the furnace exhaust stream are trapped by scrubbing in a particle separator and recycled to the melter via a pump. The off-gas passes through a condenser and a scrubbing column.

On the side of the Technology maturity, based on the DOE's TRL methodology, the IN CAN melting process has been developed by CEA for an internal application (defense waste vitrification) and can be rated a TRL 6. That means it has been tested at full scale at a prototypical demonstration level with a range of simulant in cold operation and relevant environment. The qualification for all expected conditions is quasi-complete and the design is virtually complete, engineering commissioning has started. For the decommissioning vitrification application, we can rely on all this technology development and the TRL can be rated around 4. That means that the process can be considered as a similar system demonstrated at full scale but still have to be tested with fully relevant simulant (waste, glass) to possibly finalize technology for a prototype level and reach level 7 permitting commissioning studies and full qualification to start.

Figure 11 : In-can vitrification furnace

Overview of A R&D Program

The R & D program to achieve the implementation of simplified vitrification equipment adapted to the problem of dismantling site is structured as follow (see Figure 12):

- A short first phase, to study the **feasibility**. This phase will lead to a basic design. During this first phase, a preliminary assessment of glass matrix will be achieved as well as the definition of the fundamentals of the process. A first series of tests using the existing pilot scale equipment would first approach a process flowsheet. TRL 4 to 5.

- A second phase of **development** and tests. This phase will be used to design and build a pilot scale prototype, refine glass design, start glass sustainability studies and start the engineering studies for industrial layout and nuclearization. TRL 5 to 6.

- A third phase, for process **qualification** for commercial operation, resulting in a book of final process and a summary report of qualification to support the definition of an outlet for the glass package. The pilot scale prototype will be modified if needed from phase 2 results and then use to achieve the qualification tests (nominal operations, startup/idle/shut down operations …). The launch of the studies and construction of a commercial tool could start without waiting for the complete end of the qualification phase and phase 3 would reach a TRL 7.

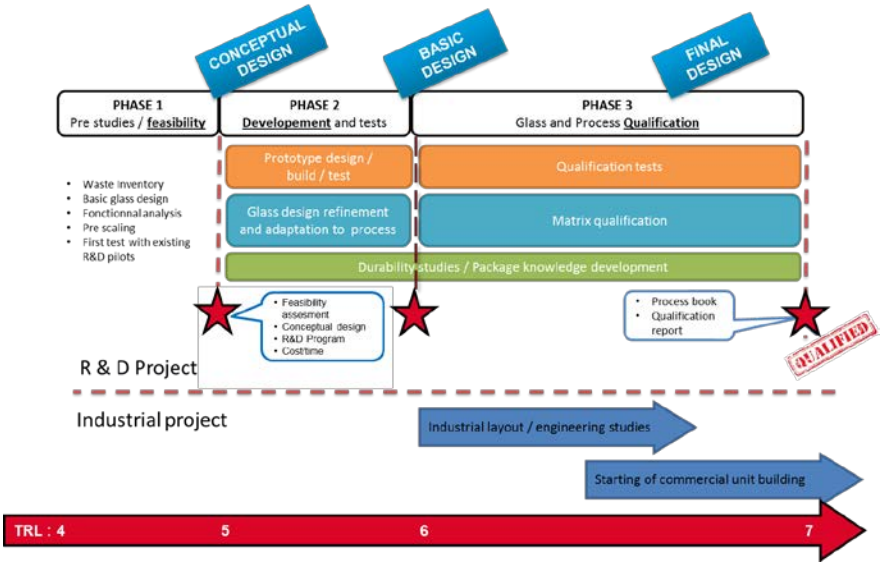


Figure 12 : Simplified R & D Structuration for a full demonstration program

CONCLUSIONS AND PERSPECTIVES

The first investigations on the FP concentration process components were performed in 2004 using the available operating equipment. The temporary storage of SPF-AVM rinsing effluents as well as the flushing operations performed prior to the definitive workshop shutdown have significantly changed the radiological inventory in the evaporator feeding tanks. The intrusive investigation campaign launched in 2010 has highlighted this severe increase of the residual source term. The methodology and the equipment specifically designed for this investigation campaign have proven to be efficient and robust. Despite the severe radiological constraints and the deposits heterogeneity, the sampling operations of tanks 71.21 A, B, C have been successful, allowing achieving a comprehensive radiological and chemical characterization program. Based on these sample analysis, the new evaluation of the workshop's mobilizable source term amounts to 22000 TBq of Cs-137, about 90% of which is located the 3 tanks 71.21 A, B, C. The high specific activity, the significant amount of actinides, the presence of solvent degradation products and the heterogeneity of the deposits raise important waste management issues. The specific Cs-137 activity ranging from 10^9 Bq/g to 10^{10} Bq/g make their incorporation into a cementious matrix technically and economically unsustainable. Despite the lack of material to complete the physical characterization, this new investigation campaign has provided us with enough valuable data to identify major technical challenges and project risks. A comprehensive detailed study and engineering program supported by R&D actions has been subsequently established. The project priority, within the coming 2 years, is to assess the feasibility of the evacuation of the HL deposits and effluents streams, including their transport, in the waste management routes previously discussed. In this strongly constrained framework, the IN-CAN melter technology, implemented on-site, could significantly mitigate project risks by:

- producing waste packages compatible with an existing on-site interim storage (SVM),
- minimizing transportation of B or C-type waste,
- accommodating waste stream polluted with organics,
- accommodating solids as well as liquid waste streams.

[1] Development of In-Can Melting Process Applied to Vitrification of High Activity Waste Solutions (HAWS): Glass characterizations and process tests results – 12442", P. Gruber et al., Waste Management Int. Conf. 2012, Phoenix USA.