Does the Perfect Waste Package Exist? Development of a Decisional Tool Addressing this Challenge-17053

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ABSTRACT

Keywords: storage, disposal, waste package, WAC

The definition of a safe, technically, economically and societally acceptable waste management strategy is highly dependent on the Waste Acceptance Criteria in each country and on the already available disposal routes. It is the reason why there is not a single solution but a combination of optimized solutions depending on the type of waste and on each country's considerations. In fact, when operating a nuclear plant or performing its decommissioning or legacy waste management, one has to consider a large waste diversity in term of natures, types, volumes and radioactivity spectra. All nuclear operators have to take-up the challenge of implementing solutions for conditioning its waste in packages suitable for transport short to long term interim storage and final disposal of it (or prepare for final disposal if the repository is not yet available). Operators have then to answer the following dilemma: either define a complete strategy packaging the waste for final disposal, or "containerizing" it temporarily, pending final disposal. The strategy is highly country dependent; in the first case, one has to take into account the available information and future orientations relative to acceptability of the packages in order to define a robust solution. The approach implemented in France requires a close dialogue between the Operators, the Safety Authority, the Administration and the stakeholders. Moreover it pushes forward whenever possible standardization, contributing to cost minimization. If the second approach has the advantage of leaving the options open, and reducing the initial investments, it inevitably requires future retrieval and re-packaging with potential evolutions/degradation of the initial waste form. It also induces the potential risk to produce additional secondary waste. As an additional drawback, it introduces uncertainties and unknowns related to the future waste management criteria and to the costs. The general idea is that, depending on context, a given route (i.e., treatment, conditioning, and transport), could be considered as efficient or not.

Assessing a waste package is certainly highly context dependent. However it should be possible to recognize and to lead an evaluation based on a number of fixed criteria, whatever the context, to be addressed when assessing the development of a final waste package in connection to the overall management route. A waste package is always a combination of a waste container and a waste form and will have to be transported one day from the production site to the final repository or to off-site interim storage facilities. AREVA's approach consists in defining an evaluation grid based on a set of criteria which aim (i) to evaluate whether the different waste treatments, conditionings, transport and packages deserve to be considered within different national contexts, and (ii) to evaluate R&D actions to be performed for improving existing solutions or developing new ones. One of the key lessons to be drawn from AREVA's experience in developing waste packages is that the perfect waste doesn't exist even though the vitrified canister is probably the most performant one. The performance of a package resulting from a compromise depends on context and is a strength combination between the waste container and the waste form. The approach is to evaluate a waste package development against different set of criteria and to draw optimization diagrams. In such diagram, a given waste package is assessed with respect to selected criteria such as waste form and waste container, stability, level of waste incorporation, integrity lifetime, secondary waste generation, transport, long term behavior, and economical effectiveness, etc. Addressing a waste form is a very key point. This evaluation grid and comparative approach will be presented through examples and comparison providing from AREVA feedback on waste package development related to complex waste management such as problematic waste or legacy waste.

INTRODUCTION

The definition of a safe, technically, economically and societally acceptable waste management strategy is highly dependent on the waste policy in each country and on the already available routes. The conditioned nuclear waste has to meet technical requirements in the waste acceptance criteria (WAC) of the existing or future management and disposal options that are unknown. The waste generator and conditioner is responsible for establishing an appropriate waste package, taking into account the national policy, available conditioning techniques, transport regulations, requirements for interim storage of waste packages, and waste acceptance criteria related to disposal facilities.

a) Whenever there is an existing authorized disposal site, then the licensee has to demonstrate how the waste meets the acceptance criteria for that disposal site.
b) Otherwise, for projects under development such as the French national deep geological repository (CIGEO), one has to supply, and substantiate as appropriate, detailed information about :
    a. the waste streams (including their origin, characteristics, inventory and quantities),
    b. the relevant devices involved for conditioning,
    c. the method used for the segregation and characterization of waste and also the actions taken to avoid dilution.

Any waste package, either existing or in development, needs a production license from the Nuclear Safety Agency (ANDRA in France).

For new designs, the Nuclear Safety Agency is continuously informed and reviews the development process. Of key importance to the project is the need for the licensee to interact at an early stage with the Nuclear Safety Agency.

There is not a single optimal solution but different conditioning solutions depending on the type of waste and on the national policy for intermediate or final disposal routes. Radioactive wastes arising from nuclear plant operations, decommissioning or legacy waste retrieval management, consist in a broad waste diversity in term of natures, types, volumes and radioactivity spectra. All nuclear operators have to take up the challenge of implementing solutions for conditioning its waste in packages suitable from intermediate interim storage transport to long term interim storage and final disposal (or prepare for final disposal if the repository is not yet available).
One of the major challenges of the nuclear waste management policies is to ensure that every waste generated has or will have a final disposal solution. For over forty years, France has opted for industrial repositories as a safe and sustainable waste management solution. The principle of disposal consists in isolating the waste from people and the environment for very long periods of time. Many other countries have chosen to set up a long-term solution based on deep disposal differing by the concept and/or the nature of the geological layer.
In France, radioactive wastes are classified in 5 waste categories as function of the level of radioactivity and the half-life of the radionuclides they contain. For each category a disposal solution route is defined. The five categories are the following: very-low-level waste (VLLW), low- and intermediate-level short-lived waste (LILW-SL), low-level long-lived waste (LLW-LL), intermediate-level long-lived waste (ILW-LL), high-level waste (HLW).

Three types of repositories has been defined or envisaged to take all French nuclear waste regardless of the level of radioactivity or the lifetime [1]. Industrial shallow and deep repositories, respectively already exist in France providing definitive solutions for VLLW and LILW-SL representing 90% of the total volume of radioactive waste generated each year in France.

Since the cost disposal increases as the depth of repositories increases, depending on the nature and radioactivity of waste the conditioning approach may be different:

- VLLW originating from dismantling, (e.g., concrete and soil), is conditioned in dedicated packages such as drums or "big bags" after sorting without specific treatments. The VLLW LLW conditioning is not a big issue, except for very low quantity of orphan waste (about 0,3% .

- The Intermediate and high-level waste need to be submitted to specific treatments before conditioning in primary packages.

Different kinds of treatment techniques on the basis of radiological, chemical and/or physical properties have been developed to allow waste handling and reduce waste volumes. Incineration, melting (e.g. scrap metal), bitumen, cementation, compaction, and vitrification (e.g., for fission products) are eloquent examples.

Volume

% Volume 2013	
30,2	VLLW
60,4	LL-SLW
6,2	LL-LLW
3	IL-LLW
0,2	HLW

Activity

Activity %	
VLLW	<0,000004 %
LL-SLW	0,02%
LL-LLW	0,01%
IL-LLW	2%
HLW	98%

Complexity

Degree of complexity	
VLLW	Low
LL-SLW	Low up to medium for orphan waste
LL-LLW	Medium
IL-LLW	Medium to high for legacy waste
HLW	High

FIG 1: Volume activity and complexity of the French National Inventory [2]

The development of these techniques lead to the development of universal packaging canisters for vitrified waste resulting from treatment of used nuclear fuel (UC-V) and compacted waste (UC-C). These canisters are considered optimized and adapted to the need.

The waste management and conditioning issues today are mainly related to ILW generated by former industrial operations such as the so called « legacy waste » often stored into temporary silos and some coming from dismantling operations. Their management, retrieval and conditioning are major technical challenges because their origin, chemical and radiological compositions are not well known. Although legacy wastes represent only about 2 - 3 % of the global inventory in France (see Figure 1), their treatment and conditioning are major issues because of the technical complexity and the cost of their management (retrieval, treatment, conditioning and packaging). Legacy wastes (ILW) are characterized by both an elevated activity of long-lived radionuclides and a broad chemical and physical variability. Sludges from radionuclide co-precipitation, powdering particles, mineral and organic adsorbent (ion exchange resins), reactive metals U, Mg, Al, and activated metals) and sometimes in mixture are some examples. In the context of end of life operations of the former recycling plant UP2-400, AREVA is responsible, for example, for the conditioning of 9000 m^3 of sludges produced by co-precipitation between 1966 and 1994, still stored in silos waiting for conditioning or others silos containing a mixture of hulls, resins, slimes or precipitates containing Pu, shearing particles, limes, powdering metallic materials. Due to technical and financial goals, the search for a convenient solution concentrates the efforts.

A methodological approach has, then, been defined to guide the design of the waste package, taking into account all the parameters and constraints and finally to find the best compromise between technical feasibility, costs and public acceptance.

HOW DEFINE THE OPTIMIZED WASTE PACKAGE?

Interdependencies of all the requirements

The waste encapsulation in acceptable waste package answering technical, economic and societal acceptance is done in a context with many uncertainties. This methodology aims at looking for the best compromise taking into account a multi-criteria approach based on safety, public acceptance and economics related to an acceptable waste package, through the whole life-cycle, from retrieval to long term disposal [3]. As shown if Figure 2, the waste package is the final product of the waste management routes and has to answer to:

- Waste package conditioning criteria
- Waste package transportation criteria
- Waste package storage criteria
- Waste package disposal criteria

The development of an acceptable conditioned waste involves different actors such as the waste producers (nuclear operators), the operators of logistic and the operator of disposal. These different actors have constraints sometimes in contradiction between them.

<u>Waste package conditioning</u>: The operator looks for the best available techniques that are as easy as possible to operate including maintenance operations which are a big challenge for a process that takes place in hot cell. It looks also for the definition of parameters easily measured to control the waste package under QA.

<u>Waste package transportation</u>: The transport requirements respect the IAEA regulation and the national safety requirements. They are often more restrictive than requirements for storage and disposal especially concerning the mechanical and fire resistance. Currently, the usual practice is to develop a cask suitable for a specific type of waste . The conditioning of wastes directly in a thick walled transportation cask is also an explored route. AREVA develops a waste package "all in one" solution able to meet the transport safety requirements. This kind solution represents a very performant waste conditioning solution in particular regarding the properties of the container. The TN-MW cask family allows a clear gain on the container performance without the need to develop a specific transportation cask for each waste container.

<u>Waste storage:</u> Safety is ensured both by the waste package itself and the confinement properties of the interim storage facilities. The characteristics required for the waste package is the mechanical resistance for handling and stacking. The waste package must ensure radioactivity confinement (gas release, swelling), and

the resistance to corrosion as a function of time. The design of the interim storage facility should provide satisfactory protection for waste packaging against deterioration (e.g., moist salt air) and adequate filters and ventilation especially when degassing the waste package.

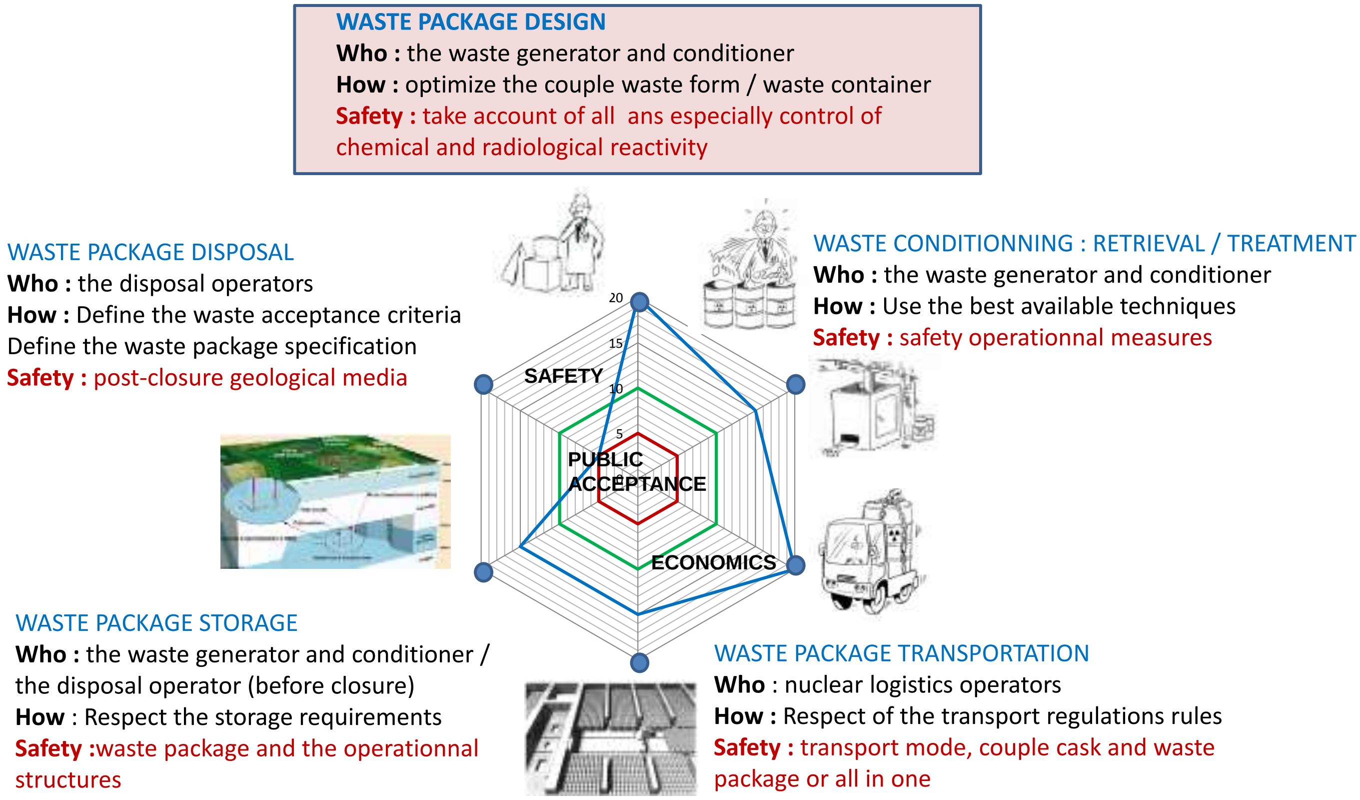


FIG 2 : Requirements for a good waste package among their life cycle

Waste disposal: Concerning the deep geological disposal, called CIGEO project, there is no formal waste acceptance criteria and waste specifications yet but the primary packages constitute the first static barrier of protection. It must ensure radionuclide confinement until, at least, the reversible phase of the long term disposal. The studies attempt to demonstrate, on one hand, the integrity of the waste container during the phase of exploitation of the final disposal (100-150 years) and on the other hand to define its behavior regarding to species release during the phase of progressive water saturation able to modify the surrounding conditions with an increase of the relative humidity within the vaults and the arrival of water in contact with the waste.

Waste form/waste container

As defined by IAEA [3,4], waste containing high activity or containing long lived radionuclides (half-life >30 years) generally requires radionuclide stabilization and must be conditioned in a container able to confine radionuclides in the disposal facility environment in order not to represent potential hazard for inadvertent intruders after closing.

Intermediate level waste belonging to this category can be divided into three categories based on stabilization and on confinement requirements:

- Heterogeneous waste not requiring stabilization
- Waste requiring stabilization and confinement due to activity or longevity of radionuclides
- Waste requiring special packaging and shielding due to handling requirements during the operational phase of the repository

The waste form refers to the physical and chemical form of the waste after treatment and conditioning resulting in a solid product prior to packaging. The primary safety requirements applied to the waste form is to confine and retain the radioactivity in the waste which usually requires processing or immobilization of the waste items (e.g., producing sludge by chemical co-precipitation). Waste forms can be homogeneous or heterogeneous consisting of various materials and objects.

The final waste package is always a combination of a waste container and a waste form. It must meet minimal technical requirements such as compressive resistance, load bearing capability, weight, resistance to impact corrosion and fire.

Characteristics of the waste form:
The waste form refers to the waste in its physical and chemical form after treatment and conditioning. The primary safety requirement applied to the waste is to confine and retain the radioactivity. Treatment and/or immobilization of the waste are, then, usually needed. The superiority of a waste form as respect to another can be defined as the extent and range of performance criteria that it is capable to satisfy.
Some waste forms may not provide high performance for one criterion, but could still be interesting by means of additional dispositions taken to compensate the poor performances. Complementary processes or additional technological and geological barriers can be added to the cask itself, at the storage or at the disposal facilities so that the waste (waste form + waste package) meet the confinement requirements. (Even though efficiency of these barriers is potentially more difficult to be demonstrated and proven than it can be on the waste form or packaging itself.). For example if the waste form is not well suited for transportation, one can add a package or an overpack to compensate or mitigate.

The larger the number of fulfilled, the more the waste form is to appear "superior". In addition, because forms are going to be used for long period of time, it is of interest that the waste form is not significantly changed through technical improvements in the future. Mature designs and forms or packaging are therefore preferred. By the way, storage or disposal designs or site selections, or public acceptance (which usually are long term activities) for such forms, could be processed with more confidence and success, when the form design is stable.

The selection of a suitable waste form has led to extensive development on matrices mainly based on cement and bitumen if we except glass dedicated to fission product solution. Extensive R&D work has been done to ensure physical and chemical compatibility between the waste and the immobilizing materials.

Unfortunately, by going all out to define a matrix, we forget the drawback of the matrices essentially linked to the reactivity between the waste, the matrix and sometimes the container.

INDUSTRIAL FEEDBACK

Feedback on waste packages

Within this context, the Universal Canister for vitrified waste (or UC-V) is especially relevant when considering issues such as criticality, safety, safeguards, high stability and high waste incorporation. Vitrified waste is the best available technology and it is a highly mature development. It is therefore likely to remain optimal for a long period of time. It is standardized and simple enough to be easily manageable, well understood and safely operated by stakeholders. The Universal Compacted Canister for metallic parts of used fuel is particularly optimized in terms of volume minimization.
The UC standard strategy provides the ability to rationalize the global waste management from receipt and storage of waste to its final disposal (see Figure 3). The selection, design and construction of the geological disposal site, as well as on-site handling and transportation operations, are indeed facilitated thanks to the standardization of the packaging with limited volumes. The resulting costs are also consequently reduced.

UC canisters can be passively stored in above ground facilities such as the "EEVLH" commissioned at the La Hague site, with a design life of 75 years. EEVLH design is basic and straightforward – with no active cooling – which provides robust, simple and comprehensive solutions for interim storage management. The concept is modular and allows for gradually expanding the overall capacity. It is flexible enough to comply with strategic changes in the future. These Universal Canisters are optimized. The waste form such as vitrification is particularly adapted to the confinement for the high activity radionuclides of the fission solutions while compaction is suited to pieces of metallic parts. Concerning the waste packages, the stainless steel universal canister answer to the container requirements including stacking and corrosion integrity.

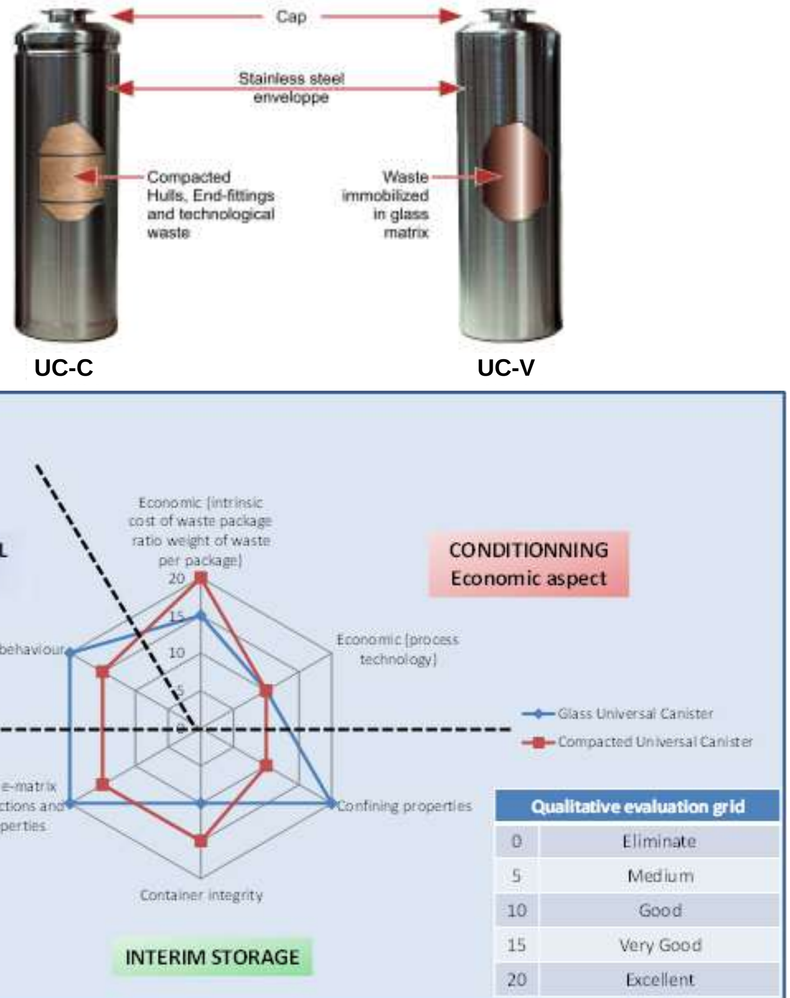


FIG 3: Multi-criteria analyses for the UCC and UCV waste packages

Feedback on matrices

It is important to test the durability of waste forms under conditions likely to be encountered in a repository setting both during operation and after closure (immersing testing, compressive strength, etc.). Furthermore, changes in waste form properties over time are likely to affect long term leaching behavior.
Typical research and development work have been done on the following topics:

- Composition selection tests for immobilizing and coating materials
- Strength testing
- Confinement properties
- Gas generation
- Degradation tests (radiation damage, corrosion)

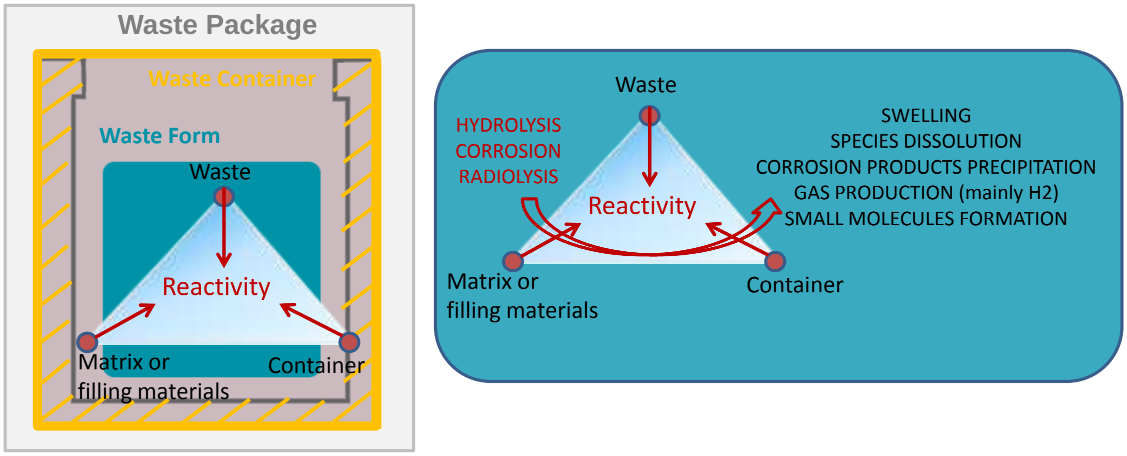


FIG 4 : Waste package : waste form/waste container and reactivity

The waste package is always a combination of a waste form and a waste container and it is submitted to the reactivity of the whole components (Figure 4). Up to now, the efforts have been mainly focused on the waste form through the development of suitable matrices such as cement or bitumen. The chemical interaction of waste, such as sludges, mineral adsorbants, powdering metallic particles, with matrix leads to different phenomena like corrosion, hydrolysis, water adsorption and swelling.

Bitumen

Conditioning of salts in a bitumen-based matrix shows drawbacks in term of radiolysis of the matrix by interaction with the radionuclides, water adsorption of salts leading to the swelling of the matrix [5] [6] [7] [8]. This process tends to be abandoned today.

Cementation

The cementation of sludges, salts, or resins requires the development of a specific formulation which is often not very robust with the chemical variability and drive to low incorporation rate (10 - 20 % in weight for homogeneous cementation in the best of cases). We note problems of instability of the matrix with phenomena of cracking due to the development of expansive phases in the case of salts or to the destabilization of system due to the propensity of resins to absorb the water [9] [10] [11] [12].

Concerning cement matrices, the interaction between the matrix and the waste can also lead to the radiolysis of the water contained in pores with production of hydrogen according to the nature of the radionuclides (alpha, beta or gamma emitters). The production of hydrogen by radiolysis and/or corrosion represents one of the major issues in management, conditioning and transport of wastes. The production of hydrogen by radiolysis can be limited by reducing the activity and, therefore, the amount of incorporated wastes. This is the reason why in certain countries such France, UK and Belgium □ and □□ activity contained in cemented wastes is limited to values as low as 1 TBq / m3 and 200 TBq / m3, respectively to insure an acceptable H2 flowrate [13] [14]. An alternative route, developed for a few years, consists in developing matrices less sensitive to the radiolysis than the classical Portland cement usually used as matrix for immobilization of radioactive waste. It is the case of geopolymeres [15] or optimized formulation of cement. This approach, although interesting, does not allow to considerably increase the incorporation rate of wastes. The same phenomenon is observed for activated metallic waste. In the presence of Mg or of Al, hydrogen production arises from the corrosion of galvanic coupling between the different metals in the presence of water contained in the pores of the hydraulic binder materials [16].

Finally, despite the numerous effort on matrices, waste packages are not much optimized in terms of incorporation of wastes, and waste form present failure regarding the interaction and the reactivity. Experience feedback shows that matrices often contribute to destabilize the total balance and chemical stability of the system. If we push forward the analysis it can be shown that the presence of free water is the common denominator to the observed phenomena of corrosion and radiolysis.

Some type of waste for which the equation is difficult to solve!

Case of the HAO Silos

Silos contain a mixture of hulls, metallic pieces, resins, slimes or precipitates containing Pu, fine particles of shearing. The general process to retrieve and condition is presented on Figure 5. Two wastes packages are identified, the first flow composed with the metallic pieces and traces of resins will be compacted in the UC-C and the second flow rate composed with the small particles will be cemented.

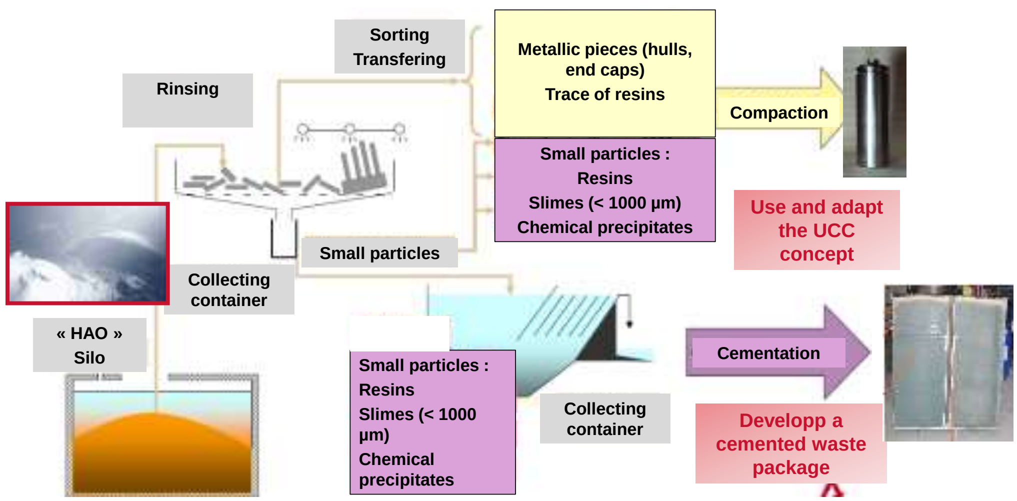


FIG 5: Conditioning approach for mixed waste in silos from former UP2-400 recycling plant in La Hague

The technical impossibility to separate the fines particles from the organic resins leads to developing a specific hydraulic binder allowing the incorporation of both in the waste. After lab experiments and test at scale, a cement binder CEM IIIC was developed and shown to have good properties. It allows an incorporation rate of about 11 weight percent with a good chemical stability, with no heating, perfect

confinement of the powdering metallic particles and good mechanical properties. This waste form satisfies the requirement of confining the small particles containing alpha emitters. But, this homogeneous matrix leads to an interaction between the fine particles and the free water contained in the hydraulic binder. A calculation tool called DOREMI [14] was established to evaluate the H2 production due to radiolysis. Based on the incorporation rate of 11 weight percent and a perfect statistical repartition of the fine particles in the global population of waste package, the evaluation of the hydrogen production leads to an average to 120 NL/year/waste package. The preliminary ANDRA limits requirements for the CIGEO project today point out a limit to about 40NL/year/waste package.
This example shows that the best confinements of these fine particles are irreconcilable with the hydrogen production except for limiting the quantity of waste. This does not appear reasonable or acceptable with regards to the cost of waste storage in the deep geological disposal.

Sludges stored in silos from effluent treatment station

9000 m3 of sludges produced between 1966 and 1994 are currently in silos. These sludge are produced by co- precipitation treatment on effluents from the former UP2-400 plant and are composed with two different salts:

- 80% of insoluble salts which trapped the radionuclides (Cs, Co, actinides)
- 20% of soluble salt such as sulfates or nitrates

Although producing sludge by chemical co-precipitation should be considered as an immobilization technique as respect to IAEA recommendations, the first explored conditioning route was to look for a matrix allowing immobilizing these sludges. The bitumen was refused by the French Authorities both because of the exothermic reactions which could eventually occur during bitumen process, and because of possible swelling due to water adsorption of salts as well as radiolysis. The immobilization of sludges in a cement-based matrix was finally abandoned after several years of R&D because of the difficulty in finding a formulation adapted to the variability of sludge compositions.

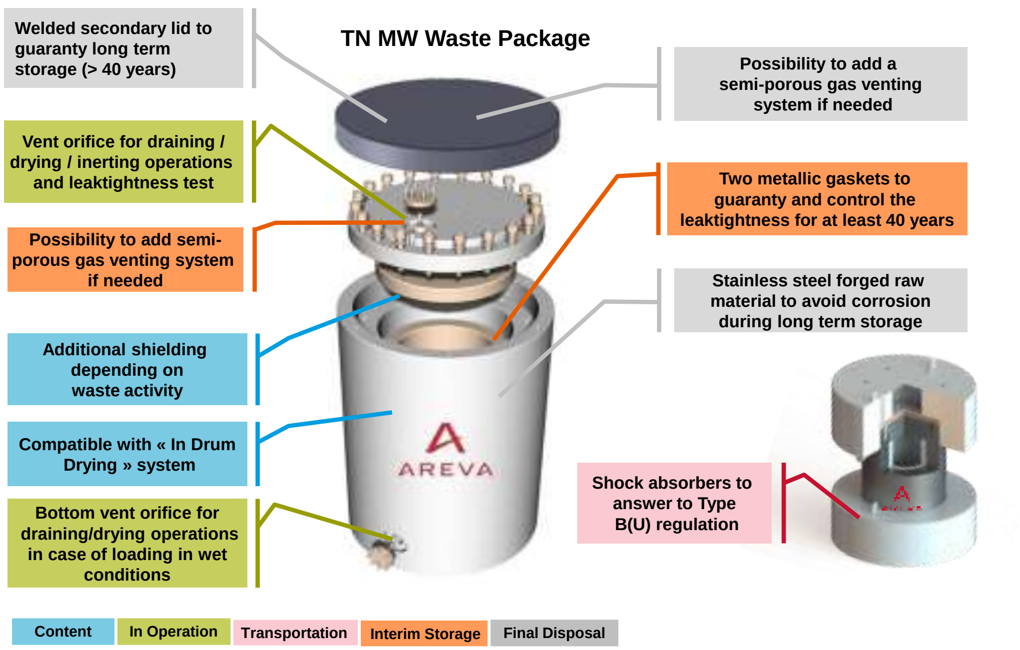
In spite of efforts led to develop matrices, some problems of reactivity remain. Feedback from experience shows that, contrarily to what is usually admitted, the use of matrices often lead to a degradation of the chemical stability of system due to the presence of water. The best way to limit the reactivity of the system is to limit or suppress the driver of reactivity, i.e., the free water, and move toward a drying system. Table 1 compares the advantages and drawback of bitumen, cement and no matrix for sludge or salts but with very strict drying.

Sludges/salt/	Conditioning process	Waste form integrity	Long term performance
Bitumen	High waste proportion ~ 40% in weight Relative low cost Operation : risk of exothermicity Fire risk to be manage	Global good confining properties Risk of swelling : water absorption, hydrogen production	Release controlled by the solubility of species with a slight slowdown of the chemical species release
Cement	Low proportion of waste ~ 10-20% in weight Relative low cost Operation: easy to operate	Global good confining properties Risk due to chemical compatibility Expansive phase formation Radiolysis and H2 production	Release controlled by the solubility of species with a slight slowdown of the chemical species release Risk due to chemical compatibility Radiolysis with H2 release
Drying with no matrix or filling material	High proportion of waste	Confining to be reported on the container. Chemical reactivity controlled by the control of free water	Release controlled by the solubility of species Chemical reactivity controlled by the control of free water (dryness content)

Table 1 : comparison of different conditioning approach for sludge and co-precipitation salts

Towards a change of paradigm

These difficulties connected to the reactivity between the waste and the matrix, phenomena of hydrolysis, of radiolysis, of corrosion as well as the low rate of incorporation of waste in particular for the homogeneous cementation led several countries to find innovative routes and change their policy of waste management by conditioning the waste directly in a transportation cask and/or storage [17], [18] [19]. This new route represents a change to the current baseline of waste conditioning and presents the benefit to simplify the operation and to reduce the volume of wastes. This technology is mainly in development for three families of waste: metallic waste, sludges from co-precipitation of radioelements, and organic ion exchange resins and/or mineral absorbers.
To answer this need, AREVA developed a waste package « All in one solution » [20] [21] designed for storage, transport and long term disposal. The diversity of typologies of wastes requires making this all in one waste package a "Swiss army knife" with in particular, a breathable system allowing the evacuation of gases if needed, a system of introduction and of filling interstitial space, and a system for drying.



This package aims to answering challenge for high value and high complexity waste

- (i) by reducing the chemical reactivity of the wastes typically resins, metallic materials by elimination of the water, common denominator at the origin of reactivity and of observed consequences (hydrolysis, radiolysis and corrosion) and by the same by increasing considerably the rates of incorporation,
- (ii) by rationalizing / standardizing packages following the example of containers standards developed for vitrified residues UC-V and compacted UC-C, and by standardizing the packaging to allow a cost cutting of transport and operation
- and with (iii) an adaptive and incremental approach allowing to answer successively the phases of storage, transport and final disposal with the same container in a progressive and reversible way.

CONCLUSION

The design of the good waste package is very tricky to define because it results from a compromise between several equations with several unknown parameters and sometimes it is possible to wonder if a solution allowing reconciling all criteria exists. The multi-criteria approach has the interest to highlight the zones of compromise and to identify the way without solution. It allows to drive the development of new approach equilibrate between the waste form and the waste container. The development of "all in one" waste package which is a very resistant waste container based on qualities requested for the transport and answering with the implementation of options such as in drum drying, breathing system, fulfill materials to the problems highlighted by the use of matrices. It proposes an early conditioning for long term storage up to final disposal, avoid the development of new concept of packages and overpack at each time and avoid the multiplication of the handling operations at each stage. It could constitute for high complexity waste, involving strong cost, and strength public acceptance, industrial, economic and societal gains.

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