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SAFETY OF LONG-TERM INTERIM DRY STORAGE OF USED NUCLEAR FUEL

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ABSTRACT

Interim storage of used nuclear fuel is a reliable solution. It provides an intermediate solution while waiting for a decision concerning disposal sites or recycling. Intermediate storage is safe as shown by important industrial feedback and the operational records. Nevertheless, this safety and reliability are well established within a domain of constraints: the respect of regulatory requirements for storage systems and a limited time, 40 years, sometimes extended.

For 25 years, interim storage systems have provided an excellent level of safety. Safety studies cover very severe accidents and natural disasters or extreme conditions: aircraft crash, fire, earthquake, cask burial, cask tip-over, fuel cladding breach. Aircraft crash testing has been achieved successfully on cask specimens.

Storage time may now have to be extended. Whatever the choice be for the management of used fuel, it will have to be transported from the storage facility to somewhere else for centralized storage, for recycling or for disposal. In the debate, two different questions are raised: the technical investigations and the safety principles.

For the assessment of long-term storage, several investigations are presently being carried out concerning the following issues: degradation process of fuel cladding, degradation of neutron poison material, degradation process of canister material, stress corrosion cracking, concrete degradation, seal degradation.

It is important for sustainability to maintain two safety principles:

- First, that there should be an *end point* to the interim storage period, and that radioactive waste shall be managed in such a way that it will *not impose undue burden on future generations,*
- Secondly, *retrievability* for safe management of used fuel

There is no guarantee that the fuel characteristics can be maintained in perpetuity. The objective of the R&D is to set a limit for which the cladding integrity is maintained. This is the first line of defence for handling, transportability and especially for safety criticality evaluations.

In the long term, R&D investigations must progress taking into account the principle that storage shall remain an intermediate step as well as the principle of retrievability of used fuel.

1- INTRODUCTION

Used nuclear fuel (UNF) management is a critical process in the nuclear fuel cycle. Numerous countries are still assessing and implementing sustainable UNF management solutions. As implementation of recycling has not yet been decided and since final disposition has not yet been resolved in many countries, wet and dry storage solutions have been deployed to support continued plant operations. However, nowadays, pools at reactors are becoming full and used fuel assemblies are being removed at the same rate as reactor discharge to interim storage. Dry storage has become increasingly prevalent as nuclear plants run out of room and options.

AREVA has developed different UNF dry storage solutions worldwide [1].

a- Canister solutions:

The NUHOMS® system, which consists of the storage of canisters in concrete modules, was initially developed for 20-year interim storage at reactor sites. The used fuel is placed in leak-tight welded canisters or dry shielding canisters (DSC) filled with inert gas, inside a massive concrete bed or horizontal storage module (HSM) outside the reactor on the site. The first barrier for the containment of used fuel assemblies is the dry shielding canister. DSCs will have to be transferred into a transportation cask for transport to centralized storage, reprocessing or final disposal.

NUHOMS® system is the leading technology used in the USA. In total, about 700 NUHOMS® systems have been loaded. NUHOMS® systems can accept BWR, PWR and VVER used fuels. A new generation of NUHOMS® systems is currently under development by AREVA with higher performance (especially higher fuel burnup).

TN NOVA ™ system is another canister system designed by AREVA. It is a system with a metal storage overpack which can be tilted vertically for storage on site. The canister is licensed for both storage and transportation.

The advantages of these solutions are modularity, passive cooling and low up front costs. However, as storage time is extended, fuel retrievability may become more challenging.

b- Dual purpose casks:

Dual purpose casks are metallic casks which can be used for both storage and transportation to and from a storage facility to avoid handling the fuel assemblies twice. They comply with the requirements of transport regulations [3]. AREVA has developed metallic dual purpose casks with more than 280 casks loaded. Dual purpose casks are in operation in the USA, Japan, Belgium, Switzerland, Germany and Italy. The capacity reaches 40 PWR used fuel assemblies or 97 BWR used fuel assemblies, and maximum 65 to 70 GWd/t burnup, cooling times of 5 to 7 years for the contents are accepted in these storage designs.

The advantages are modularity, passive cooling, and dual purpose storage and transport capability. Containment and dose are monitored during the interim storage.

As shown by industrial feedback and operational records, interim storage of UNF has been performed safely and reliably. Nevertheless, storage durations are constrained by regulatory limits. These limits are typically 20 to 60 years. Until recently, a storage period of up to 60 years was considered as 'long-term' and sufficient to consider decisions and implementation of a back-end fuel cycle and/or final waste management options. Now, it appears that storage duration may have to be extended much longer, potentially beyond one century. However, safety during the different phases seem to be quite critical: on-site interim storage, transfer and transportation operations to the final disposal site or recycling facility. Additional safety evaluations will be needed to provide a basis for the justification of interim storage solutions in the long term.

2- SAFETY PRINCIPLES

As a principle, it has been recognized that radioactive waste shall be managed in such a way that undue burden will not be imposed on future generations. Moreover, applicable and accepted principles concerning UNF management and the storage of used fuel are the principles of defence in depth and used fuel retrievability [2].

Defence in depth:

To protect the public from radioactivity hazards, containment barriers are required, including the concept of defence in depth. These containment barriers isolate the radioactive material (used fuel) from the public and the environment. They should be able to resist severe conditions with some margin. Designers have provided satisfactory solutions in terms of storage systems, canisters or packaging. In addition to the protection given by the container design, some contribution from the used fuel has always been considered as a part of defence in depth. The cladding of the used fuel rods being the first safety barrier to contain the radionuclides, its integrity must be checked during all the phases of the back-end of the fuel cycle.

Retrievability:

Used fuel retrievability is essentially a sustainability principle to select a stable and mastered system. This means that accumulation must be avoided (for example ever-increasing space, surface, volumes, waste), and obsolescence and the lack of competence must be prevented.

Future operations such as used fuel handling, transportation or unloading necessitate the maintenance of our infrastructures. During intermediate storage, activities such as surveillance, maintenance, replacement and reconditioning are or can be requested anytime. Moreover, it is necessary to maintain knowledge, competencies and trained people for all the required tasks: design, operation, checking and inspection, maintenance.

Finally, as the safety of future operations is based on the knowledge of the UNF status, fuel behavior in transport after dry storage is a key issue for the management of the back end of the nuclear fuel cycle.

3 – TECHNICAL AND SAFETY INVESTIGATIONS ON USED FUEL BEHAVIOR

The knowledge gaps have been identified by experts worldwide, for example in [5] and [6]. These gaps concern mainly severe high burnup (HBU) fuel. Higher burnup results in higher heat decay and increased fission gas release, generating higher stresses for longer periods of time in the fuel rod cladding. In addition, the longer residence time of a fuel assembly in the core to achieve a higher burnup tends to reduce the residual wall thickness through waterside corrosion further increasing stress. We have summarized the main remaining issues in Table 1.

Table 1 Data gaps on used fuel degradation modes

Used fuel rod degradation mode		Present knowledge/control	Gap/R&D program
Storage or Transport Normal conditions	Cladding creep	Degradation is limited through the temperature	In case of new alloys
	Delayed Hydride cracking	Unlikely in storage	Some investigations
	SCC corrosion	Unlikely in storage	None
	H_2 Effect Reduction of ductility	Degradation is limited if low BU	Unknown, tests required for HBU

	H_2 effect Shift in the Ductile brittle transition	Degradation is limited if low BU	Unknown, tests required for HBU
	Rod pressure He build up, pellet swelling	Unlikely in storage	None
	Irradiation damage recovery	Degradation is limited if low BU	Investigations needed for HBU
Transport Accident	Impact resistance	Tests and analysis method performed	May request additional analysis

Thermal creep is a limiting degradation mechanism for evaluating cladding integrity during dry storage. At temperatures between 300 and 400 °C currently considered at the start of dry storage, the cladding undergoes strain due to creep. Because the temperature decreases continually during dry storage, creep is not anticipated to produce significant strain after the initial storage period [9]. Under normal conditions of storage, it is now commonly agreed that creep will not cause gross rupture of the cladding and that the geometric configuration of the spent fuel will be preserved, provided that the maximum cladding temperature does not exceed 400°C.

Consideration of normal storage cladding oxidation under inert atmospheric conditions and appropriate drying procedures rules out the presence of oxygen. Therefore, no further increase in oxide layer thickness over and above the condition existing at final discharge from the reactor is expected.
Consequently, Zircaloy oxidation is not a limiting factor for cladding degradation during normal dry storage operations of LWR fuel. In addition, oxidation of M5® material is very limited [10]. Moreover, some storage systems include monitoring to detect and quickly correct abnormal conditions.

The phenomenon of cladding corrosion induced by fission-products (iodine) is unlikely because iodine is not present in a chemical form that could trigger stress corrosion cracking in dry storage.

It is agreed that for delayed hydride cracking (DHC) of LWR used fuel the critical flaw size needs to be unrealistically large. DHC has not been observed worldwide in LWR cladding.

Hydrogen effects and subsequent embrittlement of cladding is generally considered to be a possible issue in the long term, requiring evaluation and specific R&D action. While the reactor is in service, hydrogen is dissolved in fuel rod cladding made of Zircaloy. The hydrogen amount depends on the alloy, on the thermo-fluidic conditions and on the oxide thickness. For a burnup of 50 GWd/tU, the hydrogen content is 600 ppm for Zr4 (low tin), and less than 100 ppm for M5®. During reactor discharge, hydride may precipitate in the form of platelets; the observed orientation of these platelets is circumferential for Zr4 SRA (stress released annealed) or more random for Zr2 RXA (recrystallized). These hydrides have great influence on mechanical resistance of rod cladding, especially on rupture properties.

Reorientation of these hydride platelets may occur if the temperature of the drying transient is higher than the dissolution temperature and if during cooling the temperature becomes lower than the precipitation temperature. In addition, internal pressure is an important factor. Hydrogen content and hydrides affect mechanical properties for the cladding material. When Hydrogen content increases, ductility decreases, especially at low temperatures. When radial hydride fraction increases, there is also a decrease in ductility. When the fuel rod cools down, the temperature may reach the ductile brittle transition temperature (DBTT). Industrial experience has shown no embrittlement of the fuel rod cladding, but R&D investigations are underway on this matter especially for HBU fuels.

When used fuel is submitted to severe loading such as forces and accelerations during regulatory drop tests defined in [3], impact resistance needs to be evaluated. Few experiments have been done due to the difficulty in handling irradiated F/As.

The Fuel Integrity Project (FIP), performed in France and the UK by AREVA and INS, aimed to develop a methodology to evaluate, as a safety requirement, the nature and the extent of LWR fuel assembly (FA) damage during accidental dropping of a container. Test results have led to the definition of the FIP methodology [12]. Experimental knowledge was collected from the testing program, and the primary mechanical phenomena arising from a drop have been identified and quantified.

The application of the Fuel Integrity Project methodology leads to the criticality hypotheses for the safety analysis: the existence or not of fuel rod rupture, the number, the location, the associated amount of released fuel material, and the extent of fuel rod array deformation and sliding.
Tests were carried out on used fuel segments from irradiated samples of up to 40-50 GWd/tU with lateral bending of PWR and BWR rodlets until rupture, and with lateral bending of BWR rod extremity.

For the lateral drop, the slight packing down of end grids and collapse of central grids lead to limited rupture risks, whereas complete packing down of the fuel pin array in the FA central part leads to uncontrollable rupture risks.

For the axial drop, a similar analysis was carried out: for limited accelerations, the LWR end nozzle bends, and consequently fuel pin bending will occur leading to limited rupture risks. For higher loads, local plastic buckling may occur, leading to uncontrollable rupture risks.

Through analysis of the tests, the potential ruptures and the amount of fuel released upon impact were assessed showing maximum 3.5 pellets per section of broken fuel pin. These values are confirmed by other experiments [13].

4- TECHNICAL AND SAFETY INVESTIGATIONS ON STORAGE SYSTEM COMPONENTS

The degradation processes of the materials and components that need to be evaluated [8] for dry storage and/or transport systems concern:

- The canister or cask material
- The concrete
- The neutron poison materials
- The sealing system
- The coatings
- The neutron shielding materials

4-1 Degradation process of canister or cask material

Long-term issues concerning canister behavior are: Stress Corrosion Cracking (SCC), Aqueous Corrosion, and Atmospheric Corrosion. Investigations are underway for Chlorine induced SCC for canister systems in marine environments: amount of Chlorine in ambient atmosphere, assessment of deliquescence of salt, evaluation of corrosion through inspection of existing storage systems, modeling of corrosion and SCC. Mitigation techniques (stress reduction) are also being considered.

4-2 Concrete degradation

The concrete overpacks or storage modules provide radiation shielding and protection of the casks or canisters from the environment. The long-term issues concerning concrete are associated with temperatures and radiation levels: concrete chemical degradation carbonation, corrosion of embedded steel, coupled mechanisms, dry-out and thermal degradation of mechanical properties. Results from industrial experience and inspection are satisfactory.

4-3 Neutron poison material

In used fuel dry storage and transport systems, the basket generally includes neutron poison materials. As a result of extended time under applied stress at elevated temperature, neutron poison may creep. Temperature and stress can strongly influence creep behavior over long periods. Significant creep of neutron absorbers could affect criticality considerations, particularly flooding during retrieval or transportation accident scenarios. No observation of creep of neutron absorbers has been made due to control of temperatures over extended storage times.

Wet corrosion and blistering can be avoided if low porosity materials are selected. Past observed blisters are believed to be a result of water entering pores in the material during loading, leading to internal corrosion. Heating during drying and/or increasing hydrogen produced from continued corrosion over time can cause internal pressure in sealed pores to increase. This pressure can lead to blistering where the aluminum cladding plastically deforms and separates from the inner absorber material. Wet corrosion and blistering may cause dimensional changes affecting criticality considerations due to moderator displacement as well as retrievability function.

In the low porosity neutron poison materials included in AREVA dry storage systems, wet corrosion and blistering of neutron absorbers are unlikely.

Thermal aging effects, including decreases in tensile and yield strength, may occur as a result of extended time at elevated temperatures. Thermal aging effects on neutron absorbers are not operative if the design temperature is limited.

The embrittlement of neutron absorbers due to radiation exposure is unlikely for the selected materials during the storage period. Only slight decreases of elongation were experienced in the reactor radiation environment due to damage caused by fast neutrons and neutron flux anticipated during storage period are orders of magnitude lower [8].

To summarize, degradation of the properties and safety functions of the neutron poison materials during the dry storage period is unlikely for AREVA dry storage basket designs and the selected materials.

4-4 Cask materials or components

a- Seal degradation

For metal casks, the main containment issue is the resistance of the sealing system, especially metal seals. Long-term issues are the corrosion of bolts and the corrosion of metal seals. The behavior of these components has been studied under storage conditions with satisfactory results by the CEA (France) and the CRIEPI (Japan). These studies cover long-term resistance (leak tightness tests) of metal seals with Al or Ag outer jackets. BAM corrosion tests on such seals have also shown good resistance.

b- Coatings

Polymeric coatings are currently used on external surfaces of Dual Purpose casks. These coatings have three different functions:

- Ease the heat dissipation by increasing emissive thermal exchanges
- Protect the steel against corrosion, eventually in addition to metallic thermal spray
- Ease the decontamination of the external surfaces after loading in the reactor pool

After cask loading, thermo-oxidation of the coating is the main degradation phenomenon. The evolution of the functional properties (hardness, adhesion, emissivity …) should be studied during thermal ageing, and should be correlated with chemical transformations in the paint binder for understanding paint degradation.

At AREVA, R&D studies showed that after long-term storage under normal conditions the epoxy paints used on AREVA casks are still emissive and free of defects. This latter characteristic ensures corrosion protection (in addition to thermal spray coating) and ease of decontamination.

c- Neutron shielding

In-service neutron shielding material property changes are caused by irradiation or thermal oxidation processes. To our knowledge, however, experimental results show that oxidation is by far the most predominant process. Therefore, our recent work has been concentrated on the oxidative mechanisms of our thermoset matrix-based neutron shieldings. Our methodology [14],[15],[16] can be detailed as follows:

- Accelerated aging tests are carried out on neutron shielding samples at both various temperatures and O_2 partial pressure. Then, characterization of aged samples is performed.
- O_2 permeation tests carried out in order to estimate both O_2 diffusivity and solubility into neutron shielding films.
- To investigate the long-term properties, a non-empirical model is applied. To this end, oxidation kinetics is coupled with oxygen diffusion. The model very confidently simulates weight losses (which are then converted into hydrogen atoms loss) and oxidation profiles.

Simulation data agree with experimental results for all conditions under study. This allowed us to predict in-service hydrogen atom loss over decades in the neutron shielding materials.

5- CONCLUSION

Currently R&D studies are progressing satisfactorily to demonstrate the safety of dry storage as an intermediate step. Storage system and component degradation is very small or unlikely. R&D is being performed on several knowledge gaps. One of the most significant is the question of chlorine induced stress corrosion cracking of stainless steel in the case of storage systems in marine environments. Used fuel behavior is also a key issue for the safety of transportation after long-term dry storage. R&D studies are needed, especially concerning the embrittlement of the high burnup fuel cladding, linked to hydride reorientation. These studies will provide the necessary basis to justify the safety of interim storage and transportation in the long term.

It is important for sustainability to maintain the necessity of an *end point* to the interim storage, and the retrievability of used fuel for further use, either disposal or reprocessing.

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