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Spent Fuel Performance Assessment and Research: Final Report of a Coordinated Research Project (SPAR-II)

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Spent Fuel Performance Assessment and Research

Final Report of a Coordinated Research Project (SPAR-II)

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FOREWORD

As of the beginning of 2008, approximately 300 000 tonnes of heavy metal (t HM) in spent fuel had been discharged from nuclear power reactors. Only about one third of this spent fuel has been reprocessed, and thus approximately 200 000 t HM in spent fuel is currently stored in various storage facilities. Most of this fuel is under water, but dry storage is becoming a widely used technology, with more than 25 000 t HM currently stored in this manner in various countries. Spent fuel storage capacity needs will continue to increase in the future, and some of this fuel will have to be stored for 50 years or longer before it is reprocessed or undergoes final disposal. As a result, interim storage for long periods of time will continue to be a key technology for all Member States.

A number of countries are planning or have already initiated research programmes on spent fuel storage performance, or are developing a high confidence database that evaluates the storage of spent fuel for extremely long periods of time to determine the consequences for disposal. In addition there is increased interest on the part of Member States in future nuclear cycles and possibilities to recycle nuclear fuel. This interest would imply the long term storage of current fuel before it could be reprocessed and recycled.

Spent fuel storage technology (particularly dry storage) is undergoing an evolution, new fuel and material design changes are coming on stream and burnups are steadily increasing. These improvements call for new studies and potential adaptations of the storage technologies currently used.

As a consequence, in 2004 the IAEA continued the coordinated research project (CRP) on spent fuel performance assessment and research (SPAR-II) to collect and exchange the spent fuel storage experience of the participating countries; to build a comprehensive international database supporting the licensing of present and future technologies; to carry out research work to evaluate and justify the storage of spent fuel for long periods of time (more than 50 years); and to assist in defining how the requirements for spent fuel storage and for the whole back end of the fuel cycle are connected.

The present publication is based on the results of investigations undertaken in the participating countries. The draft document was prepared and discussed during the last Technical Meeting, held in Budapest, Hungary, 2–6 June 2008. The report provides an overview of technical issues related to wet and dry spent fuel storage and summarizes the objectives and major findings of research carried out within the framework of the CRP. The results reported here reflect the particular interests and research capabilities of the participating countries. It is not the aim of this report to provide a complete overview of the current status of the spent fuel storage industry.

The IAEA gratefully acknowledges the contribution of the CRP participants and the consultants who participated in the drafting and reviewing of this publication. The IAEA staff members responsible for this publication were W. Danker, Z. Lovasic and Ho Jin Ryu of the Division of Nuclear Fuel Cycle and Waste Technology.

EDITORIAL NOTE

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SUMMARY

As storage of spent fuel has become a key technology in spent fuel management, wet and dry storage have become mature technologies and continue to demonstrate good performance. Increased spent fuel storage capacity in combination with longer storage durations will be needed over the foreseeable future as many countries have delayed their decision on spent fuel disposal or reprocessing. Extended spent fuel storage is, and will remain, an important activity for all countries with nuclear power programmes. A number of countries are planning or have already initiated research programmes on spent fuel storage performance, and there is a continuing benefit in exchanging spent fuel storage experience of the Member States in order to build a comprehensive technology knowledge base.

Potential degradation mechanisms that may affect cladding integrity during wet storage are uniform corrosion, pitting, galvanic, and microbiologically-influenced corrosion. Potential degradation mechanisms that may affect cladding integrity during dry storage and subsequent handling and transportation operations are air oxidation, thermal creep, stress corrosion cracking (SCC), delayed hydride cracking (DHC), hydride re-orientation, hydrogen migration and re-distribution. Investigations carried out so far indicate that from the degradation mechanisms that may affect the integrity of spent fuel assembly/bundle structure during interim storage, hydride re-orientation has the potential to impair the ability of the cladding to effectively withstand potentially adverse mechanical challenges resulting from handling or transportation accidents.

Fuel integrity issues are related to the definition and criteria of fuel integrity, failure classification, packaging and retrieval of damaged fuel and transport of damaged fuel assemblies. Various monitoring technologies have been developed and used to confirm the continued spent fuel integrity during storage or to provide an early indication of developing conditions which would impact on the long-term spent fuel integrity.

Vacuum drying and hot gas recirculation drying are employed in preparation leading to dry storage. The drying process is of importance to the dry storage systems as it prevents the presence of any water remnants which could potentially facilitate some degradation mechanisms during storage. Temperature increase during drying has to be controlled to minimize the potential for hydride re-orientation.

In addition to research on spent fuel assembly integrity, research activities on the behaviours of storage facility components including reinforced concrete, neutron absorbers used in storage racks and baskets, neutron shielding materials, metal gaskets, polymer seal, stainless steel and carbon steel are also being conducted.

As a conclusion, it remains important to continue to study and share results regarding fuel and materials behaviour in storage. In particular, changes in fuel and material design, and the increase in discharge burnup require continued research on spent fuel performance in long-term storage and post-storage transportation.

1. INTRODUCTION AND OBJECTIVES

Dry storage technology is undergoing a rapid evolution. New fuel and material design changes are coming on stream and target burnup is steadily increasing. Average fuel assembly burnup can currently approach 60 GWd/t U in LWRs, 33 GWd/t U in gas reactors, and 10 GWd/t U in CANDU reactors. Increased burnup, particularly for LWRs, results in increased cladding corrosion and fission gases leading to increased cladding stress. The International Spent Fuel Conferences held by the IAEA in Vienna in 2003 and 2006 confirmed that these changes require continued analyses and potential adaptations of the storage technologies currently used, especially taking into account that spent fuel may have to be stored for extremely long periods of time; e.g. for up to 100 years and beyond.

In the future, demands for spent fuel storage capacity will continue to increase, with some countries planning to store for up to 100 years or more prior to subsequent operations. As a result, interim storage for long periods of time will continue to be a key consideration for a number of Member States.

The first dry storage facilities have reached the end of their initial licensed period. As final spent fuel repositories are neither in operation nor under construction, licenses for these interim storage facilities have to be extended or renewed. The United States investigated a storage cask and its contents after 15 years of storage to obtain data to support license extensions. Other countries will likely follow this example and also perform some investigation and/or monitoring.

The international conferences on spent fuel from power reactors mentioned above recommended that the IAEA should provide assistance in the evaluation and research of the long term behaviour of fuel and storage components in order to realize the anticipated long storage periods and stressed the importance of international collaboration on specific issues. The President of the 2006 Spent Fuel Conference concluded that while ‘specialists expressed confidence in the technical development of storage facilities and containers, in order to satisfy long term needs, it is clearly necessary for more research and development on the fuel behaviour in dry storage.’

Presently, spent fuel is mostly stored wet in at-reactor (AR) or away-from-reactor (AFR) facilities. Because many AR pools are approaching their full capacity, even after extensive re-racking, storage of spent fuel under dry and inert atmosphere is being used increasingly.

The need for further investigation of spent fuel performance is further expanding as today’s reliance on higher burnup fuels as well as on mixed-oxide fuel, to generate electricity at a competitive cost increases. Given the much higher decay heat levels from these fuels, wet storage will remain the preferred approach for interim storage during the first decade, after discharge. After sufficient decay (‘cooling’), and especially for long storage durations (up to 300 years under some scenarios), dry storage under inert conditions or in air, becomes the preferred alternative, given the passive nature of dry storage systems.

This publication builds on the IAEA Spent Fuel Performance Assessment and Research (SPAR) Coordinated Research Project (CRP) and reports on a continuation phase of the same name but identified as SPAR-II. The SPAR CRP was a continuation of the previous BEFAST-I to III (Behaviour of spent Fuel Assemblies in Storage) CRPs.

The overall objective of the SPAR-II CRP was to develop a technical knowledge base on long term storage of power reactor spent fuel through evaluation of operating experience and research by participating Member States. Specific research objectives included:

- Fuel and materials performance evaluation under wet and dry storage;
- Monitoring programmes of spent fuel storage facilities;
- Collection and exchange of relevant spent fuel storage experience of the participating countries.

The second phase of the IAEA Spent Fuel Performance Assessment and Research (SPAR-II) CRP (2004-2008) involved the European Commission as well as 13 other organizations from 12 countries: Argentina (joined later), Canada, France, Germany, Hungary, Japan (2), Republic of Korea (only for a limited period), Slovakia, Spain, Sweden, UK, and USA. Sweden and the UK participated in the programme as active observers. Three research coordination meetings were held during the course of the SPAR-II CRP: the first in June 2005 in Karlsruhe, Germany; the second in November 2006 in Tokyo, Japan; and the third in June 2008 in Hungary.

2. HISTORY OF THE BEFAST AND SPAR COORDINATED RESEARCH PROJECTS

Extended spent fuel storage is, and will remain, an important activity for all countries with nuclear power programmes because fuel after its discharge from the reactor is required to be stored before reprocessing or final disposal. The storage period is highly dependent upon the individual national strategies to close the nuclear fuel cycle, but generic questions related to spent fuel storage are common to all nuclear programmes.

The first phase of the BEFAST programme (1981-1986) involved 12 organizations from 11 countries: Austria, Canada, Czechoslovakia (CSSR), Finland, Federal Republic of Germany (FRG), German Democratic Republic (GDR), Hungary, Japan, Sweden, USA, and USSR. A subsequent programme, BEFAST-II, implemented during the years 1986-1991, involved organizations from 12 countries: (Argentina, Canada, FRG, Finland, GDR, Hungary, Italy, Republic of Korea, Japan, UK, USA, and USSR). BEFAST-III, implemented during the years 1991-1996 involved 15 organizations from 12 countries: (Canada, Finland, France, Germany, Hungary, Japan, Republic of Korea, Russian Federation, Slovakia, Spain, UK and USA). There was also an observer from Sweden.

During the three BEFAST Co-ordinated Research Projects (CRPs), the participating countries contributed their R&D results on fundamental questions of spent fuel storage. The reports of the CRPs have been published as IAEA-TECDOCs [1].

Towards the end of the BEFAST-III programme, it became apparent that the R&D component of the programme was decreasing steadily; more emphasis was being placed on the operation and implementation of storage technology. The storage technology (particularly dry storage) was undergoing a rapid evolution: new fuel and material design changes were coming on stream and target burnup were steadily increasing. With the increased burnup came higher fission gas and fission product inventories, increased sheath (cladding) strains and increased cladding hydriding and oxidation. Because of all the new parameters that have surfaced during the course of BEFAST-III, a subsequent programme was proposed to address the effects of these new parameters on long-term storage and to determine their consequences on disposal.

The first phase of the IAEA Spent Fuel Performance Assessment and Research (SPAR) CRP (1997-2002) involved 11 organizations from 10 countries: Canada (2), France, Germany, Hungary, Japan, Republic of Korea, Russia, Spain, Sweden, UK, and USA. Sweden participated in the programme as an observer [4].

Major topics for both wet and dry storage during all five Coordinated Research Projects are summarized in Table 1. It records detailed objectives and the shift in emphasis during the various phases of the programmes between 1981 and 2008.

Table 1. RESEARCH SUBJECTS IN PAST AND CURRENT CRPS.

Long-term behaviour	B-I	B-II	B-III	SPAR	SPAR-II	Surveillance	B-I	B-II	B-III	SPAR	SPAR-II	Facilities and operation	B-I	B-II	B-III	SPAR	SPAR-II
Material aspect (cladding & components)	X			X	X	Monitoring; Wet + Dry -Environment - Components - Fuel assemblies - Workers' dose rate			X	X	X	Capacity enhancement - High density racks - Re-racking - Double tiering - Doped coolant - Rod consolidation		X			
Degradation mechanisms and models	X	X		X	X	Fuel conditions - Operational - Fabrication - Technology - Defected fuel rods and assemblies		X	X	X	X	Changing modes Wet – Dry		X	X	X	X
Validation - experiment - experience		X	X	X	X	Different reactor types		X	X	X	X	Handling of heavily damaged fuel		X	X		X
												System performance	X	X	X	X	X

B=BEFAST, I-III

3. SYSTEM CONTEXT FOR STORAGE

3.1. INTERIM NATURE OF STORAGE

Member States continue to deploy increased storage capacities successfully, with both wet and dry approaches considered to be fully mature technologies. Storage is considered to be an interim stage in the backend of the nuclear fuel cycle, with the understanding that spent fuel management operations including storage and/or reprocessing are ultimately followed by final disposal. As one perspective on the interim nature of spent fuel storage, the IAEA published a position paper on spent fuel storage in 2003 [5].

Several options for the ultimate management of spent fuel discharged from nuclear power plants are being implemented or under consideration. At the present time, the two leading options are:

- Direct disposal of the spent fuel in a geologic repository (once-through cycle), and
- Reprocessing of the spent fuel, recycling of the reprocessed plutonium and uranium, and final disposal of the wastes from the reprocessing operations.

However, delays in opening geologic repositories in most countries or in implementing reprocessing in some countries mean that increased spent fuel storage capacity in combination with longer storage durations will be needed over the foreseeable future. Worldwide, spent fuel production rate, now at about 10 500 t HM, is expected to increase to ~11 500 t HM by 2010. Because less than a third of the fuel inventory is reprocessed, ~8 000 t HM/year on average will need to be placed into interim storage facilities.

In 2005, the US Nuclear Regulatory Commission granted a 40-year extension to an interim spent fuel storage installation operating under a license for an initial 20-year period using dry storage technologies. The USNRC staff paper supporting this action identified advantages and disadvantages of several options for extension. One of the disadvantages identified by NRC staff for the 40-year extension highlighted the resultant 60-year storage duration as re-defining the term ‘interim storage’.

The trend toward extended storage durations was highlighted in the summary findings of the President of the June 2006 IAEA spent fuel conference as follows: ‘Most spent fuel storage systems were designed for short term application pending reprocessing or disposal. The time period requirements for storage systems have been extended in most countries because of the unavailability of disposal facilities. In some countries, new facilities have been built for the purpose; in others, the use of existing facilities is being extended for longer periods.’

Wet and dry storage continues to demonstrate good performance and provide system flexibility pending further progress in the back end of the nuclear fuel cycle. As spent fuel storage quantities and durations increase, the storage component in the nuclear fuel cycle represents an important, flexible ‘spring’ in the system, accommodating changes and delays related to interfacing components. It therefore remains important to confirming the continuing viability of this spring by continuing to study and share results regarding fuel and materials behaviour in storage through efforts such as the Spent Fuel Performance Assessment and Research (SPAR-II) CRP.

3.2. NATIONAL APPROACHES TO STORAGE[1]

(1) Canada

The spent fuel from Canadian CANDU reactors typically remains in wet storage for ~ten years before being placed in dry storage, with the total accumulation expected to reach ~3 500 000 fuel bundles. They have an active program for researching storage effects for both wet and dry storage facilities, but the number of interrogations tend to be a very small percentage of their large inventory.

The Nuclear Waste Management Organization was established in 2002 with the requirement to assess three spent fuel management options (long-term at-reactor storage, long-term centralized storage, and deep geologic disposal) within three years. The Report by NWMO assessed the benefits, risks, and costs for these options but went further by recommending an alternative approach, ‘adaptive phased management’. This approach consists of preparing for centralized management (phase 1), central storage and technology demonstration (phase 2), and long term containment, isolation, and monitoring (phase 3).

(2) France

France is a country in which reprocessing of spent nuclear fuel has been chosen for the back end policy and has henceforth less need of interim storage systems. This has been confirmed in the national plan included in the new radioactive waste management law passed in June 2006. In order to reduce the radioactive waste stream and to recycle nuclear material, the reprocessing route for spent nuclear fuels in France is confirmed.

Spent fuel from NPPs is transported to La Hague reprocessing facility. There, spent fuel assemblies are unloaded from transport casks and stored in the La Hague pools for cooling before being reprocessed. The 6 pools of La Hague represent a capacity of 18 000 metric tonnes of spent fuel, including 14 000 metric tonnes for EDF.

When the NPP pools are full, storage can be achieved either in a new facility or in modular systems such as metal casks. Spent fuel can be stored on the site of the NPP or transported to a storage site away from the reactor site. When spent fuel final disposal is considered, it is necessary to foresee transport operations anyway. That is why in the context of storage, the question of transport is always to be considered.

(3) Germany

Due to the Act for the Regulated Termination of the Commercial Use of Nuclear Power, a spent fuel management concept was promulgated in 2002 saying that interim storage shall bridge the time period until a final repository will be available.

Presently, storage experience is available from centralized installations, such as Ahaus and Gorleben. In future, new experience will be obtained from the decentralized interim storage installations at the sites of the nuclear power plants. These on-site facilities are mainly storage buildings with duration of license of 40 years. They were already licensed and have been

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[1] Because the following statements in Section 3.2 were made as of 31 Dec. 2008, the present tense means the status of 2008.

under construction since then. Interim storage areas with 5-8 years duration of license are used until the on-site facilities will become operational.

Fuel integrity assessment under dry storage conditions is featured by as standard licensing methodology based on thermal creep degradation where uniform plastic strain and stress are limited. To cope with further developments that may challenge current limits, further creep tests are being performed on high burn-up materials in hot cells under largely realistic long term conditions, i.e. 400°C and 130 MPa cladding stress.

(4) Hungary

The foundation of the policy and distribution of the responsibilities for the radioactive waste management and its final solution was updated in the Act CXVI on Atomic Energy (Act) adopted by the Parliament on December 10, 1996.

Under the Act, the Director General of the HAEA was authorized by the Government to set up the Public Agency for the Radioactive Waste Management (PURAM) which tasks are, among others, to deal with final disposal and interim storage of spent fuel and radioactive waste. PURAM has been in operation since 1998.

The Central Nuclear Financial Fund was established through the Order of the Minister in 1997. The Fund is administrated by HAEA and will be used to finance the construction and operation of any waste facilities and the decommissioning of nuclear facilities.

Work on research, development and demonstration for deep geological disposal of spent fuel and high-level, or long lived radioactive waste has started.

The situation in Hungary can be summarized as follows:

- 4 Units of Paks Nuclear Power Plants in operation;
- Nuclear power supplies about 40% of the country’s electricity production;
- A modular vault dry storage (MVDS) facility was commissioned, presently it has 11 vaults for 4 950 FAs (approx. 600 tU);
- Extension of the MVDS with further 5 vaults starts this year;
- Preparation of arrangements plans, technical plans, and licensing documentation for an Underground Research Laboratory for HLW disposal is in progress.

(5) Japan

At the end of 2007, 55 commercial nuclear power units were operating in Japan, with a total electric generation capacity of 49 467 MWe. Nuclear power generation is 30.5% of total electric power generation, with two power plants currently under construction and eleven plants in preparation for construction. All sixteen nuclear power plant sites have storage pools for spent fuel. The total amount of spent fuel stored at these sites was approximately 12 140 t HM as of September 2007, compared to a capacity of 19 000 t HM. Various measures for additional storage are being evaluated, including site-wide sharing of pools, pool re-racking and expansion, and dry cask storage. Japan is pursuing an active program to research the behaviour of fuel and storage components as the storage duration extends.

(6) Slovakia

Slovakia has operated altogether six nuclear units with nuclear reactors of VVER-440 type. Four units are installed in Jaslovské Bohunice (EBO 1-4, called NPP V1 and NPP V2) and two of them at Mochovce (SE-EMO 1-2). EBO 1 has been definitely shut down on 31 December 2006, EBO 2 were shut down on 31 December 2008.

Nuclear power plant A1 located at Jaslovské Bohunice was designed to use natural uranium as fuel in a heavy water reactor cooled with carbon dioxide (HWGCR – 150MW). This power plant was shut down in 1977 after an accident (INES 4) and it is in the first stage of decommissioning at present. The spent fuel from this plant was transported to the Russian Federation based on a valid contract.

An interim spent fuel storage is in operation in Jaslovské Bohunice since 1987, where the project of seismic resistance and storage capacity increase has been implemented.

(7) Spain

The spent fuel managed in Spain comes from the operation of the country's nine nuclear reactors, located at the seven sites that house its nuclear power plants: José Cabrera, Santa María de Garoña, Almaraz I and II, Ascó I and II, Cofrentes, Vandellós II and Trillo.

Eight dual-purpose casks (168 assemblies) were housed in the dry cask storage facility at the Trillo site by the end of 2004. A centralized modular vault storage facility (ATC) is planned for 2010 and targets a design life of 100 years and a capacity of ~20 000 assemblies (~7 000 t HM). Their SPAR-II research plans address high burnup fuel, investigations of hydrogen migration in fuel cladding, and dry storage operational experience.

(8) Sweden

A central interim storage facility for spent nuclear fuel, CLAB, has been built and been in operation since 1985. Over 4 000 tonnes of spent fuel are presently stored at CLAB. The work on research, development and demonstration for deep geological disposal of spent fuel has been intensive for more than 20 years. It has now reached such maturity that key decisions concerning design and siting of an encapsulation plant and a final repository are possible.

The situation in Sweden can be summarized as follows:

- Site investigations for the Final Repository for SNF in progress on two locations
- Application for the Encapsulation Plant mid 2006
- Decision in 2008 where to locate the Final Repository for SNF
- Application for the Final Repository for SNF 2011
- Start of operation for the Encapsulation Plant and the Final Repository 2023.

(9) UK

In the UK there are three basic types of nuclear power plant in operation; Gas Cooled Reactor (natural uranium magnesium non-oxidising clad fuel) -'Magnox Reactor', Advanced Gas Reactor (AGR) and one Pressurized Water Reactor (PWR).

Up to the formation of the Nuclear Decommissioning Authority (NDA) in April 2005, (under the Energy Act 2004, to take strategic responsibility for the UK's nuclear legacy), the decision on the back end policy adopted for spent fuel management was up to the power generators. The policies adopted prior to 2004 for individual fuel types were:

- Magnox (Sellafield Limited)
    - Closed Cycle (Magnox fuel is programmed to be reprocessed by end 2012)
- PWR (British Energy)
    - Open Cycle (Long term storage at reactor(currently within the capability of the existing AR pool) pending direct disposal (>2075))
- AGR (Managed by Sellafield Limited on behalf of British Energy)
    - Closed Cycle (Reprocess AGR fuel until 2010/11)
    - Open Cycle (Non-reprocessed AGR fuel will be stored wet in existing facilities (Thorp Receipt and Storage) pending direct disposal (>2075).

Since April 2005, the responsibility for the management of spent nuclear fuel at Sellafield, Magnox Reactor Stations, and former UKAEA sites has transferred to the NDA.

The NDA's approach has been to establish a national review of the longer term management options for spent fuel involving all relevant stakeholders and taking account of the outcome of the CoRWM process and subsequent decisions by the Government.

(10) USA

An application for a construction license at the Yucca Mountain site was submitted by the U.S. Department of Energy (DOE) to the U.S. Nuclear Regulatory Commission (NRC) on June 3, 2008. As part of the licence application, the DOE has re-introduced the multi-purpose canister (MPC) concept, now referred to as the Transportation, Aging, and Disposal (TAD) concept, in order to simplify handling of spent fuel at the sub-surface Yucca Mountain facilities. In the meantime, accumulation of spent fuel plus fuel presently generating power in the US reactors is approaching the *legal* capacity of Yucca Mountain. Delays related to Yucca Mountain and projections for a significant expansion of nuclear generation of electricity, coupled with the announcement of the Global Nuclear Energy Partnership (GNEP) by the Bush Administration in February 2006, have spawned a vigorous debate about the potential benefits and drawbacks of closing the nuclear fuel cycle.

With regard to interim storage, R&D programs have been mostly driven by regulatory issues centred on management of high-burnup fuel and extension of dry storage licenses beyond their initial 20-year duration. Results from some of these R&D programs are already apparent. For example, the NRC has extended the dry storage license at two different sites for an additional 40 years resulting in anticipated interim storage of up to 60 years. Also, dry storage of high-burnup fuel is no longer considered to be an unresolved regulatory issue; however, transportation of high-burnup fuel remains largely unresolved. With regard to the latter, work has included the development of models to (1) predict zirconium hydrides precipitation and reorientation in heavily corroded, high fluence Zircaloy cladding; (2) define failure criteria for claddings with both circumferential and radial hydrides under handling or transportation accident conditions; and (3) characterize the dynamic loading on the fuel rods as a result of an accidental drop of a spent fuel cask. Results obtained so far indicate that high-burnup spent fuel rods with Zircaloy cladding can retain high resistance to severe load challenges such as the dynamic forces resulting from cask drop accidents. Mechanical properties of advanced zirconium-based claddings are being investigated for conformance within the acceptable performance envelope of Zircaloy claddings.

4. WET STORAGE

Pool storage in the SPAR-II countries accounts for 130 000 t HM of spent fuel as of January 1, 2008. The largest quantities of spent fuel in wet storage reside in the US (~48 200 t HM) and Canada (29 600 t HM). Most of the spent fuel is in facilities located at reactor (AR) sites. The largest away-from-reactor (AFR) wet storage facilities are in Sweden (CLAB) or associated with the reprocessing sites of La Hague (France), Rokkasho Mura (Japan), and Sellafield (United Kingdom).

The experience in wet storing spent nuclear fuel now spans some 50 years. Wet storage continues to dominate as the primary method for storing spent nuclear fuel; >80% of all spent fuel is wet stored. The benefits provided by this technology are mainly associated with cooling efficiency and shielding. Also, it facilitates safeguards and one-off fuel inspection/examination exercises. Despite the emergence of dry storage technologies, new pools and extensions to existing wet storage pools have been initiated or completed in France, Germany, Japan, Sweden, UK and United States.

Reactors have been burning MOX-fuel since the early 1970s. Currently its usage is exclusive to Europe where there are 91 reactors with part-core MOX loading. Spent MOX behaviour in wet storage is similar to UO_2 fuel apart from the higher end-of-life neutron radiation levels and associated heat of decay. For these reasons significantly longer wet storage is required to achieve the comparable radiation and decay heat levels of UO_2 spent fuel prior to further processing (interim dry storage, reprocessing or direct disposal).

4.1. SPENT FUEL STORAGE EXPERIENCE

4.1.1. General performance

For primary barrier or containment purposes, cladding corrosion is the factor of most interest in wet storage. However, retention of fuel assembly structure integrity also becomes an important factor when retrieval is taken into consideration. A detailed review of the degradation mechanisms of various fuel types under wet storage conditions is given in Chapter 7.

For zirconium alloy clad fuel, data exist for continuous pool storage of greater than 50 years. These data indicate cladding corrosion to be extremely low (1×10^{-6} μm/a) and, therefore, corrosion is not viewed to be the time-limiting factor for prolonged wet spent fuel storage; even under poor pool chemistry conditions.

For stainless steel clad fuels, continuous storage experience of 32 years (LWR) and 38 years (AGR) exists. Although the general cladding corrosion rates for these fuels are significantly higher than for zirconium-based alloys (at ~0.1 μm/a), general corrosion is not a time limiting factor for the storage durations currently envisaged (up to 100 years). For AGR fuel, particular attention to pool water chemistry is required as parts of the fuel stringer become sensitized during reactor operation; see Chapter 7 for further information.

Magnesium alloy clad fuel is particularly susceptible to cladding corrosion under wet storage conditions. Although a protective magnesium hydroxide film is initially formed, the presence of any aggressive ions in the water promotes the dissolution of the protective oxide film and leaves the cladding open to pitting attack. For this reason, Magnox fuel is stored in dosed pool water and storage duration tends to be limited; normally <5 years.

In general, the negative impact on spent fuel and wet storage systems may be caused by the adverse pool chemistry conditions due to poor chemistry control or deterioration of the fuel storage pool structure.

4.1.2. Wet storage experience in the participating countries

The following paragraphs provide examples of the application of wet storage in the countries participating in the CRP.

(1) Canada

Canada has been storing research reactor fuel under water since the 1950s. CANDU power reactor fuel has been stored in water pools at the reactor sites for about 45 years. Currently, Canadian utilities are transferring used fuel into dry storage after a wet-storage period of about seven to ten years.

Used CANDU fuel is wet-stored in Irradiated Fuel Bays (IFB). IFBs are water pools, which are lined either with stainless steel or fiberglass reinforced epoxy compound, and equipped with purification systems.

CANDU (natural uranium) fuel has a discharge burnup of approximately 8 GWd/t U. The fuel is Zircaloy-4 clad. Experience with wet-storage has been excellent.

A program to demonstrate the feasibility of storing used CANDU fuel under water was started at AECL's Chalk River Laboratories in 1977 (see also [4]). Various fuel elements, some with intentional through-wall cladding defects, were initially stored and subsequently examined after 3–17 years. The conclusion from the examination was that intact and defective used fuel showed no deterioration and that extrapolation of the data indicated that a 50 year target storage time could be safely exceeded [6].

(2) Eastern-European Countries (Hungary, Slovakia)

Hungary has AR pools at each reactor, while Slovakia has an AFR wet storage facility (capacity 1 694 t HM – 14 112FAs) beside the AR pools.

The behaviour of the Zr1Nb clad WWER fuel in storage has been good. Over ~40 years experience of wet stored fuel exists, there have been no known fuel cladding failures as a result of wet storage; under normal operating conditions.

Corrosion studies have been conducted on Zr1Nb alloy cladding specimen being cut from WWER spent fuel assemblies of different burnup and cooling times. Results showed that burnup has a much more significant influence on the cladding plasticity than storage duration.

For WWER-440 fuel, the measured oxide film thickness is distributed uniformly along the total cladding; its average value was 0.01 mm. In the location of the spacer grids, a local increase in the oxide film thickness was observed, with an additional value of 0.005 mm [7].

(3) France

Before reprocessing in France, spent fuels are stored in medium-term buffer storages. At La Hague facility, the fuels are stored in the pools. The total surface of the pools is 5 500 m^2 and the depth of water is 9m. Spent fuels are stored in special baskets designed for handling.

There are 2 types of baskets that can accommodate either 4 PWR fuel assemblies or 16 BWR fuel assemblies. The geometry and presence of neutron poison materials are used in order to guarantee sub criticality. Damaged fuels are transported and handled in bottles before storage in the pools.

The advantages of wet storage are: first, the evacuation of residual heat of spent fuels, as water is a good heat transfer medium and the second, the protection of operators and visitors from radioactive material: water provides dose rate attenuation and containment.

In the pools at La Hague, the water is cooled by heat exchangers connected to a close cooling loop. The heat exchangers are submerged in the pools. The heat is dissipated by external dry cooling towers. The radiochemical properties of the pool water are controlled continuously by anion/cation ion-exchangers (cartridge-type mixed-bed exchangers, ionic ‘Nymphea’ concept).

Since the beginning, 30 years ago, of the wet storage at La Hague, no failure or degradation of spent nuclear fuel has been detected, it continues to show a good performance.

(4) Germany

In Germany, a little less than 3 540 t HM spent fuel is presently stored in at-reactor (AR) pools. This number might change a little with time. It was decided that spent fuel will be stored in dry storage casks at every reactor site (including Obrigheim). The central storage sites in Ahaus, Gorleben or Greifswald are dedicated to special dry storage purposes. Therefore, in the future, the amount of spent fuel in AR wet storage will decrease.

Wet storage technology has been used for Zircaloy-clad fuel since decades. No limiting effects have ever been found. This also holds for high burnup fuel with new corrosion resistant structural materials.

(5) Japan

There are 55 commercial nuclear power units (32 BWRs including 4 ABWRs, 23 PWRs) operating in Japan. Two units (1 ABWR, 1 PWR) are under construction. Eleven units (8 ABWR, 1 BWR, 2 APWR) are in preparation for construction. All sites have spent fuel storage pools. The cumulative amount of spent LWR fuel discharged from the reactors is approximately 23 000 t HM. Approximately 8 500 t HM of this amount have been shipped to domestic and overseas reprocessing plants, and the remainder is stored at reactor sites. Most of these have been stored in pools, without problem. As of December 2007, 2435 t U of spent fuels were transported to the storage facility of the Rokkasho Reprocessing Plant and about 308 t U of them were used in the test runs [8]. The main specifications of this facility are as follows:

- Maximum storage capacity:1 500 t U for BWR spent fuel and 1 500 t U for PWR spent fuel
- Number of pools: 3(1: exclusive-use for BWR fuels, 1: exclusive-use for PWR fuels, 1: common-use for both BWR/PWR fuels)
- Maximum fuel enrichment before irradiation for receipt: 5.0wt%
- Conditions of receiving spent fuel: Residual enrichment: less than 3.5wt%
- Cooling time: over 1 year before receipt
- Burnup : 55 GWd/t U (max.), 45 GWd/t U (average)

The total controlled-capacity, which excludes full core and reserve capacity, available for spent fuel storage at all LWR NPP sites is about 19 000 t HM. Due to the storage amount of spent fuel being approached to the controlled capacity being approached at a number of NPPs, and to meet future demands, re-racking (AR), expansion or modification to common-use for two or more reactors has been undertaken beside installing some dry storage casks.

(6) Spain

Spent fuel has been wet stored in Spain since 1970. In view of the forthcoming saturation of the capacity of these pools, the original storage racks were progressively replaced with more compact units during the 1990s. All wet fuel storage is AR, and storage experience has been good, apart from an occasional top nozzle separation of early manufactured Westinghouse PWR fuel assemblies (see US contribution later). All fuel assemblies in storage have been moved at least once, when the AR pools were re-racked, without any problems occurring. As of January 2008, 3 580 t HM of spent fuel was stored at the NPPs.

(7) Sweden

A central storage facility for spent nuclear fuel, CLAB, has been built and been in operation since 1985 with the current storage capacity of 8 000 t HM.

As of January 2008, the storage records for CLAB are as follows:

- 4 700 t HM are stored,
- Spent fuel maximum burnup is 53 GW·d/t U,
- MOX fuel is stored,
- Average collective dose is about 25 man·mSv/year,
- Release of radioactive nuclides to the environment is negligible,
- 22 years of successful operation,
- More than 1 600 transport casks with spent fuel has been received and processed.

In addition, about 650 t HM was stored at the plants as of January 2008.

(8) United Kingdom

The wet storage of fuel discharged from Magnox and AGR NPPs can be conveniently divided into two phases. The first covers short-term storage at the reactor and the second covers the further interim storage away from reactor at Sellafield, prior to reprocessing. The only exceptions to the above are British Energy’s Sizewell B PWR reactor, where all spent fuel is currently stored at reactor wet.

The experience of wet storing commercial spent nuclear fuel on the Sellafield site spans some 45 years. There are four operational AFR storage pools at Sellafield: Fuel Handling Plant (Magnox/AGR), AGR Storage Pond (AGR), LWR Storage Pond (LWR/PIE), and Thorp Receipt & Storage (LWR/AGR). These pools store fuel from the first- and second-generation UK reactors, Magnox and AGR, and also irradiated fuel from LWR reactors, mostly from BNFL’s overseas customers. As of January 1, 2009, 6 000 Mg spent fuel is stored in the Sellafield pools.

The main differences between fuel storage at Sellafield and other facilities are:

- Fuel storage is based upon containerized systems (Magnox and AGR containers, Multi-element bottles for LWR). These facilitate:
  - Isolation of the fuel from the bulk pool water,
  - The chemistry within the container can be different from the bulk pool water,
  - The spread of corrosion products to the storage facility is minimized.
- A variety of pool chemistries are utilized, depending on the properties of the fuel being stored.
- Apart from a few special transports, there are no individual fuel element handling operations, during storage.
- The pools are purged with fresh water; resultant effluent is either treated in the Site Ion Exchange Plant, or filtered and sentenced prior to sea discharge.

(9) United States of America

Spent fuel storage experience from the civilian reactor programme spans some 45 years. Apart from the General Electric AFR Morris facility in Illinois, all wet storage is AR. The current storage capacity is around 60 000 t HM. As of January 2008, 47 500 tonnes were stored in the pools of the reactors, and 670 t HM was in the GE Morris facility.

In general, the performance of wet storage systems has been excellent, with only occasional operational problems or incidents. In particular, top nozzle separations in older PWR Westinghouse assemblies occurred at the North Anna, Millstone, Farley, and Prairie Island sites. The degradation mechanism was identified as intergranular stress corrosion cracking of sensitized 304 stainless steel. Design and materials changes were implemented in later designs and procedural controls have been implemented for managing spent fuel assemblies that may be susceptible to nozzle separation.

4.1.3. Research activities related to wet storage

(1) France

Measurement of characteristics of fuels in wet storage has shown no degradation. Safety studies are thoroughly carried out to validate the storage concept. Loss of coolant accident is considered: fuel behaviour at high temperature and dry conditions has been studied, to validate the emergency response plan. Especially, microstructural changes in the cladding, stress-strain curves, oxidation phenomena kinetics were studied.

(2) United Kingdom

Studies have been initiated to evaluate alternative AGR fuel corrosion inhibitors to sodium hydroxide (see section 1.2.1). The purpose of this research is to address materials compatibility issues, mainly associated with the co-storage of AGR fuel with aluminum based components, to facilitate the bulk storage of AGR fuel in Sellafield’s Thorp Receipt & Storage facility.

From a short list of ten potential inhibitors four (demineralized water, sodium nitrate, sodium silicate and sodium silicate dosed sodium hydroxide) were studied using an inter-granular attack sensor, to simulate sensitized AGR cladding, in combination with the electrochemical technique zero resistance ammetry (ZRA). Because of the inconclusive nature of the ZRA testing of demineralized water, this was tested further using the metal loss technique Field

Signature Method (FSM) and was discounted. Sodium nitrate has since been taken forward for active testing with high burn-up AGR brace materials to confirm its suitability for storage deployment.

4.2. POOL WATER CHEMISTRY

The role of water in spent pool storage is to:

- Facilitate heat removal from the spent fuel,
- Act as a biological shield,
- Maintain fuel cladding integrity,
- Facilitate spent fuel visual inspection.

To achieve these goals, water quality has to be optimized for the fuel being stored. Water chemistry must also consider criticality control and microbial control. Factors that may be affected include water chemistry, water quality control, and water temperature.

Operating experience gained since the 1950s shows:

- The ingress of aggressive ions such as chloride should be minimized,
- Where pool water quality has been maintained, spent fuel storage performance for all types of fuel has been excellent,
- Optimum pool water chemistry is linked to the fuel being stored,
- Apart from some experience with water-filled, canned RMBK fuel, the general experience with defected fuel is that the defect does not propagate during prolonged storage.

4.2.1. Water chemistry

The objectives of controlling water pool chemistry are to:

- minimize corrosion of metal surfaces (e.g., fuel cladding, stainless steel pool liners, storage modules and handling tools),
- minimize the concentration of radionuclides in the water, and as a result reduce radiation fields and airborne contaminants in the pool area, and
- maintain clarity of the pool water for ease of in-pool fuel handling.

Chemical specifications have been developed to monitor quality of the water chemistry in the pools. Specifications are also used for application of corrective action levels in the case of deviation in the water chemistry.

For CANDU fuels, which use Zircaloy-4 for cladding material, water chemistry specification is based on experience gained over 40 years in Canada. Spent fuel is stored in AR pools in dematerialized water. Typical specifications include: water conductivity maintained below 200 μS/m, turbidity below 1 FTU (Formazin Turbidity Unit) and pH in the rage 5.5-9. In part, conductivity is controlled by keeping the chloride level low (< 0.5 mg/kg).

The storage of Magnox fuels requires particular attention in controlling pool water chemistry. Although magnesium alloy cladding dissolves in pure water by the following reaction:

$$Mg + 2H_2O \rightarrow Mg(OH)_2 + H_2,$$

a passive/protective magnesium hydroxide film is formed. This protective film, however, is also susceptible to dissolution from acidic species dissolved in the water:

$$Mg(OH)_2 + 2H^+ \leftrightarrow Mg^{2+} + 2H_2O$$

It is therefore important to minimize ions that would lead to cladding degradation; i.e., chloride, carbonate, and sulphate. To overcome these effects, Magnox fuel is stored in high purity caustic-dosed water to a minimum pH of 11.4. Away from reactors, these conditions are increased to pH 13.0 inside the storage containers. Fuel has been satisfactorily stored for up to 5 years at pH ≥11.4 by dosing with sodium hydroxide and keeping $[Cl^- + SO_4^{2-}]$ <1 mg/kg.

Apart from a very small amount of older stainless steel clad fuel (three NPPs), all LWR's use Zircaloy cladding. Spent fuel is stored in open structures (such as racks or baskets used worldwide), or in sealed containers (such as the Multi Element Bottle used in UK). Water purity is controlled to suppress conditions that might lead to a corrosive environment for spent fuel cladding and structures.

The AR pools for PWR reactors are filled with borated water, while demineralized water is used for BWR reactors as well as for AFR wet storage installations serving both Light Water Reactor types. Depending on the countries that operate LWRs, water chemistry for storage installations may be quite different. However, some general parameters can be defined: pH requirement is 4.5–5.5 and chloride and fluoride concentrations have to be limited to 0.1–0.15 ppm. In Japan, the pH requirement for BWR stations is 5.3–7.5; chloride and fluoride concentrations are limited to 0.5 ppm, while each operating value is generally maintained below 0.05 ppm.

For some storage pools that use Boraflex neutron poison material, the increasing concentration of silica may become a concern. The silica results from the degradation of the silicon rubber polymer and the release of crystalline silica present as filler material in the Boraflex.

Because pool water is evaporating due to decay heat, the boron concentration correspondingly increases in PWR facilities. As a result, measures are taken to bring the concentration back to the prescribed values.

WWER fuel with Zr1Nb alloy cladding is stored in borated water in the AR pools. Boric acid concentration is maintained within 12–16 g/kg; concentrations of halides (Cl^- and F^-) are kept below 0.1 μg/kg; pH varies in the 4.3–6.5 range. AFR pools are filled with demineralised water. Halide concentrations are kept below 0.15 mg/kg; pH values are within the 5.5-7.0 range.

4.2.2. Pool water activity

The main sources of pool water activity are from the leaching of activated corrosion products (commonly known as 'crud') adhered to the fuel, and from fuel or fission product species being released through cladding defects.

Water activity has to be maintained at a reasonably low level to limit dose to the operators and to minimize adsorption by in-pool structural components and ancillary equipment, as a means of facilitating final decommissioning activities.

Fuel crud is the common name given to the residues deposited on fuel assemblies as a result of reactions of the coolant with the primary coolant circuit components. It is generally observed that fuel from gas-cooled reactors has minor deposits, and PWRs are cleaner than BWRs. This is by virtue of the differences in materials used in construction and the chemistry of the cooling circuits.

The radionuclide content of fuel crud mainly comprises of cooling circuit corrosion products that have been neutron-activated. In some cases fission products may be present in trace amounts where in-reactor cladding defects have been present.

The main sources of suspended solids are from particulates associated with the flask if pool discharged, from crud dislodged from individual fuel assemblies, transfers to storage, movement of the fuel, or open fuel baskets in storage. Flask-associated particulates include crud released from the fuel (thermal shock) when water is introduced to the flask internals, if shipped dry; dirt on the flask externals; and entrained particulates from the flask liner.

In the case of open storage bays, airborne particulates are also introduced.

(1) BWR

In Sweden, crud removed from BWR fuel originating from the Ringhals-1 NPP has been analysed since 1978. This experience shows that the amount of deposited material is more or less linear with the residence time of the fuel in the reactor. The composition of the crud is feedwater water chemistry dependent.

Upon fuel discharge from reactor, typically the principal nuclides present are ^{54}Mn, ^{55}Fe, ^{58}Co and ^{60}Co, with ^{55}Fe being the dominant species. Due to the longer half-lives of ^{55}Fe and ^{60}Co, only these remain above background levels in 15-year cooled fuel. In the long-term (>100 years), the main activation species remaining is ^{63}Ni, which is present only in trace amount at reactor discharge.

Studies indicate BWR crud to be hematite in origin. Typical metal content is ~87 wt% Fe, in the form of red hematite, ~2 wt% Cu, ~4.4 wt% Zn, ~3.3 wt% Ni, and ~2.2 wt% Mn [9].

(2) PWR

The major components of PWR crud are reported to be spinels of the nickel-substituted ferrite ($Ni_xFe_{3-x}O_4$ with $0 \ll x \leq 1$) and magnetite (Fe_3O_4) types. Compounds of NiO, SiO_2, and Cr_2O_3, plus elemental Ni have also been identified. Typical metal content is about 78 wt% Fe, 20 wt% Ni, and 2 wt% other metals (mainly Cr, with Co at 0.03–0.11 wt%).

Magnetite and spinel species are slightly soluble in borated waters. As a result, PWR cruds dissolve over time, leaving fuel surfaces relatively clean after a sufficiently long wet storage period [10].

(3) Gas-Cooled Reactors (Magnox/AGR)

Fuel arisings from gas-cooled reactors mainly suffer from carbacious crud deposits from the carbon dioxide coolant reactions with inhibitors/graphite decomposition.

4.2.3. Water treatment

Water treatment normally includes a mechanical treatment to remove the solid materials contained in the pool, in conjunction with a chemical treatment to extract both radioactive and non-radioactive chemical species dissolved in the pool water.

Mechanical treatment of the bulk pool water is generally performed by filters (pre-coated sand or mechanical), while chemical treatment is realized with ion exchangers (cationic and anionic resin types are used). In some cases, ion exchange is preceded by neutralization. Generally only single bed organic ion exchange resins are regenerated when saturated. The resultant concentrate may include a boron recycling step, followed typically by evaporation and final encapsulation for disposal. Mixed bed filters and inorganic exchanger resins are also used. In these cases, there is no regeneration phase and the saturated beds are disposed of directly after encapsulation.

The build-up of particulates on the pool floor and walls is removed mechanically by in-pool cleaners. A variety of designs have been deployed from modified leisure pool cleaners, simple suction devices to purpose-built two-stage cleaners, coarse and fine (cyclone) filters.

In the case of ion exchange there are three types of systems in operation. These include: out of pool ion exchange columns, ion exchange floated on top of a pre-coated mechanical filter and in-pool water treatment units; examples of the latter include cartridge systems and the combined ion exchange/cooling Nymphea system (France).

Special attention is also required to avoid the growth of microbiological species that can reduce water clarity or even lead to microbial attack of storage materials. The main factors in limiting biological growth are to minimize the introduction of nutrients (especially phosphates), intensity of lighting in the storage area, and temperature.

Where bacterial growth does initiate, treatments have varied form the use of biocides to full-scale mechanical cleaning and collection of the bio growth. Operating pools at high pH also prevents biological growth.

4.2.4. Water temperature

Water temperature requirements may vary with the specific plant design and with the type of storage installation (AR or AFR). For AR pools, the temperature limits normally are about 45°C. For AFR storage installations, spent-fuel cooling-time is greater, water temperature is lower and does not exceed 40°C in normal conditions. In Canada, AR pool temperatures are typically in the range 20–30°C.

Normal temperature limits are usually based on operating considerations, such as personnel occupation requirements and equipment operating limits, rather than fuel corrosion. Other considerations that make it preferable to operate pools at the lowest practicable temperature include lowering the release rate of radionuclides from defective fuels, minimizing bacterial or microbial growth, and lowering the humidity level in the storage area.

In particular, the transfer of activity from the fuels to the storage water has been shown to double with each 10ºC-rise in temperature. It is, therefore, advantageous to keep pool water temperature as low as reasonably possible, thereby delaying release, and taking advantage of the decay of the activity while it is fixed to the fuel surface. The load on the waste treatment systems becomes correspondingly lower [11].

Recently, modeling work has been undertaken which adds support to a temperature dependent ^{137}Cs release rate. Cesium release rate was modeled in relation to temperature changes within the bulk pool water for a known inventory of leaking AGR fuel elements during the replacement of the pool cooling system.

This compares with previous data derived at CLAB; over the temperature range 28 to 36ºC the activity level reached a new equilibrium 2.1 times higher, where the AGR activity release model predicts a 1.8 times increase in the ^{137}Cs release rate for the same temperature.

4.3. POOL COMPONENTS AND MATERIALS

The choice of pool components and materials is dependent upon the type of fuel being stored, cost, and to facilitate final pool decommissioning. The latter property has come to the fore as experience in both in-pool performance and final pool decommissioning has been gained. This has led in most cases to the almost exclusive usage of stainless steel for spent fuel storage structures, heat exchangers, piping and fuel handling equipment.

4.3.1. Pool lining

There are two main methods of treating the internals of the reinforced concrete pool structure to make it watertight, that is to line with welded sheets of stainless steel or to coat with a water resistant paint system.

All LWR AR storage pools are stainless steel lined, CANDU AR pools are stainless steel or epoxy lined, and Magnox pools use epoxy/paint lining. All AFR storage installations are stainless steel lined (France, Finland, Germany, Japan, Sweden), with the exception of UK AFR storage pools where most are only lined at the wind/water line and the rest is painted.

In case of concrete pools coated with epoxy (Canada), the concrete employed has been shown to have negligible corrosive ion leaching and permeability to water. However, the cumulative dose rate on the epoxy has to be limited to prevent epoxy degradation. Measurable changes in epoxy liner properties have been observed after 1 MGy radiation dose. With the extension of the station operating life, this dose limit could be exceeded at some point. Since 1988, some extent of radiation-induced deterioration has been observed at the Pickering station, without any damage on the liner envelope. In the case when water temperature has been maintained higher than 32°C on a regular basis, a risk of minor damage to the coated concrete walls has been observed at the Pickering station in Canada. If epoxy liner radiation-induced deterioration continues, there is a possibility that water may eventually contact the structural concrete. Thus, a programme to investigate the long-term effect of water on concrete was considered.

To resolve the issue of epoxy/paint degradation in Magnox pools, alternative paint systems have been developed and deployed in the UK.

4.3.2. Storage racks

The designs/methods used for spent fuel pool storage can be fuel/facility type, and operator dependent. LWR (AR) fuel is typically stored in open top floor mounted borated stainless steel racks. In the case of AFR fuel interim storage, prior to reprocessing, the storage systems are designed to facilitate the bulk movement of fuel and as such tend to have a reduced capacity than can be achieved AR by the fixed high-density rack systems. In the UK, the storage systems mainly comprise the ‘basket’ used in transportation; i.e. the Magnox & AGR skips and the multi-element bottle (LWR). In Canada, fuel may be stored in modules or baskets that can be loaded directly into dry storage structures.

The materials of construction are primarily borated stainless steel, or stainless steel, usually in combination with a neutron absorbing material; for example Boral, Boraflex or Cadminox. One exception is the Magnox skip, where painted mild steel is used due to concerns with respect to the potential for electro-coupling between the fuel cladding and stainless steel. CANDU modules are also stainless steel, but not borated because of the lack of criticality concerns. The only other material that has found some use for structural components is aluminium; mainly for its corrosion resistance in deionized water when passivated.

In Sweden, CLAB, the spent fuel is stored in 'storage canisters' that contain 25 BWR, or 9 PWR fuel assemblies. The compartments in the storage canisters are made of boron alloyed stainless steel.

5. DRY STORAGE

5.1. INTRODUCTION

Almost 25 years of favourable experience exists with the dry storage of spent power reactor fuel and about 40 years with the dry storage of research reactor fuel. Dry storage experience exists with fuel from a variety of reactor types (CANDU, HWR, PWR, BWR, WWER, Magnox and HTGR). Since its conception, dry storage of spent fuel has evolved into a wide variety of systems. Examples of these are concrete canisters (Canada), steel-lined concrete containers or casks (Canada, Germany, Japan, Spain, USA), and concrete vault-like structures (Canada, France, Hungary, UK, USA)[1-3,12]. At the present time, almost all countries participating in SPAR-II (Canada, France, Germany, Hungary, Japan, Spain, United Kingdom and the United States) are engaged in various dry storage technologies. Almost all participating countries are also actively pursuing a dry storage research and development programme. So far, the results of the research indicate that fuel can be stored safely under the present conditions for many decades.

As regards licensing conditions for the dry storage facilities, different trends and licensing periods have been implemented throughout the SPAR countries as shown in the following Table 2.

TABLE 2. DRY STORAGE FACILITY LICENSING CONDITIONS IN VARIOUS COUNTRIES

Country	Initial licence period	Renewal period
Canada	5 years	5 years
Germany	40 years	none
Hungary	10 years	10 years
Japan	Not limited (similar to the reactor site licence)	N/A
Spain	20 years	20 years
United Kingdom	Not limited (covered under the reactor site licence)	N/A
USA	20 years	20 years*

* The first two Independent Spent Fuel Storage Installations (Surry and H.B. Robinson) applying for an extension of the original licensing periods received a 40-year renewal period. Rulemaking has been initiated to change the initial and renewal license periods to 40 years each.

Dry storage has become a mature technology and the quantities being placed into dry storage are increasing significantly. The inventory of spent fuel in dry storage in the above countries, as of 1 January 2008, was about 24 500 t HM.

In Canada, dry storage facilities are in operation or under construction at all 7 reactor sites. As of 1 January 2008, the total inventory of spent fuel in dry storage was about 8850 t HM.

Also, in the USA, as many of the spent fuel storage pools reach their capacity, dry storage is a significant factor in the utilities` spent fuel storage strategy. As of 1 January 2008, there are 41 dry storage facilities located at commercial reactor sites, with 10 200 t HM fuel in storage. Additionally about 2400 t HM is stored in casks at the Government sites.

As of 1 January 2008, there are 17 AFR dry storage facilities in Germany, with 2 390 t HM in storage.

A helium storage environment is used in most dry storage systems. Air as well as helium is used in Canada (helium by OPG, air by AECL). A combination of CO_2 and air is used in the dry storage of Magnox fuel in the UK. Nitrogen is used in the MVDS in Hungary and at Ft. St. Vrain (USA).

Dry storage of spent LWR fuel in an inert atmosphere is licensed dependent on burnup and type of cask for temperatures up to 410^{o}C in Germany, 400^{o}C (and possibly higher than 400^{o}C for low-burnup fuel) in the USA and Spain [13,14]. Dry storage in nitrogen is licensed for a temperature of 410^{o}C in Hungary for Zr/Nb clad fuel. Dry storage is licensed in Canada for temperatures up to 160^{o}C for storage in air. In Canada, dry storage is also licensed for temperatures of up to 360°C in helium.

Most of the fuel in dry storage is clad with a zirconium alloy (Zry-2, Zry-4, Zr2.5Nb and Zr1Nb); however, dry storage experience also exists for magnesium and aluminium clad fuels.

Average burnups of spent fuel presently in dry storage range from 4.5 to 33.5 GWd/t U, while the maximum burnups range from 7.5 to 50 GWd/t U. However, there is an almost universal tendency towards increasing the discharge burnup of the fuel elements. In Germany, for instance, average discharge burnups for PWR fuel assemblies have increased from 35 GWd/t U in 1983 to 50 GWd/t U in 1998, and a value of 65 GWd/t U is achieved presently. In the USA and other SPAR countries, most of the spent fuel assemblies are being, or will be in the very near future, discharged with burnup in excess of 45 GWd/t U. However, most presently licensed storage systems have burnup limitations of in the range from 45 to 65 GWd/t U, for LWR fuel.

5.2. DRY STORAGE STATUS AND EXPERIENCE

Dry storage experience in the countries participating in SPAR CRP as of January 1, 2008 in the SPAR countries is shown in Table 3.

TABLE 3. AMOUNT OF SPENT FUEL IN DRY STORAGE IN VARIOUS COUNTRIES

Country	**t HM in dry storage (As of January 1, 2008)**	**t HM in dry storage (Forecast by Jan 2015)**
Canada	8850**	17,500
France	50	50
Germany	2290	1 700
Hungary	600	800
Japan	213***	*
Spain	79	220
United Kingdom	-	-
USA	12 600**	~20 000

* Forecast not available.

** Government and commercial facilities

*** As of August 1, 2008

(1) Canada

Atomic Energy of Canada Limited (AECL) has been storing spent fuel from its research and demonstration reactors in a dry environment at the Chalk River Laboratories (CRL) and at Whiteshell Laboratories for decades. AECL has also designed reinforced concrete canisters which have been used since the mid 1970s to store fuel from several NPPs. More recently, AECL developed the MACSTOR modular dry storage system for CANDU fuel. These are reinforced concrete modular structures that store spent fuel inside metal containers. MACSTOR systems are used at Gentilly-2 (Canada) and Cernavoda (Romania).

Ontario Power Generation (OPG) uses Dry Storage Containers of its own design. They are transportable containers built of reinforced concrete, encased in (inner and outer) steel liners, which each hold about 7.7 t HM (384 CANDU fuel bundles). OPG has been operating a dry storage facility at the Pickering site since 1995, and at the Bruce site since 2003, and is building a facility at the Darlington site.

(2) France

Dry storage has been developed in France at the CASCAD facility for spent fuel that does not require prompt reprocessing. As of 1 January, 2008, approximately 50 t HM of heavy-water reactor fuel from the decommissioned EL4 Brennilis reactor had been stored in the vault at the CASCAD facility at Cadarache. The maximum capacity of the facility is about 100 t HM. Spent fuel is canisterized at the reactor (within a dry cell). Canisters, vacuum dried and filled with helium, are transferred to the dry-storage installation. Canisters are stored in storage wells ventilated by natural convection.

(3) Germany

Germany’s revised energy act, dated 2002, prohibits the shipment of spent nuclear fuel to reprocessing plants and restricts its disposal to a final repository. To comply with this law and to ensure further nuclear plant operation, the reactor operators decided to construct on-site facilities for dry cask storage, to keep spent fuel assemblies until a final repository is available. Decentralized interim storage facilities for spent fuel assemblies have been licensed under atomic law, constructed and commissioned at all twelve sites with operating nuclear power plants. There are three technical concepts of on-site storage facilities: Storage buildings, the storage tunnel, and interim storage areas. Whereas storage buildings and the tunnel are envisaged for a license duration of 40 years, the interim storage areas were mainly foreseen for a licensing duration of 5-8 years to bridge the time until the storage buildings will be commissioned. These temporary storage facilities used in the past years as an interim solution have in the meantime all been emptied again, and the casks have been transferred to the corresponding on-site interim storage facilities.

The on-site interim storage facilities hold 1 614 t HM, and 92 t HM LWR fuel assemblies in storage casks in the central interim storage facilities at Ahaus and Gorleben. 583 Mg HM of WWER fuel assemblies from Rheinsberg and Greifswald are likewise stored in casks at the ZLN interim storage facility in Lubmin near Greifswald. A total of 6 675 t HM of spent fuel assemblies from the nuclear power plants have already been shipped abroad either for reprocessing or for permanent storage there.

The central interim storage facilities Gorleben and Ahaus (licensed for CASTOR V/19 and V/52) remain in operation, but no new spent LWR fuel will be transferred to the site. Since 2005, Gorleben is only dedicated to the storage of vitrified HLW.

A basis for cask licensing is the integrity of the spent fuel structure and fuel rod cladding structure and the exclusion of systematic fuel rod cladding failures [15]. A systematic failure of the rod cladding in a non-corrosive environment is avoided by limiting stress and strain, whereby material's properties have to be considered.

(4) Hungary

A Modular Dry Vault Storage (MVDS) started operation at Paks to store spent WWER-440 fuel. The construction started in March 1995, the facility was commissioned in 1997 and the first 450 assemblies, filling one vault were loaded in 1997. Successive vaults were filled during the later years, presently the first 9 vaults are full, while the 10th vault is filled partially. As of January 1 2008, total of 5107 assemblies were in the MVDS, which has 11 vaults constructed and further 5 vaults under construction.

(5) Japan

Dry storage facilities were constructed at the Fukushima Daiichi Nuclear Power Station of Tokyo Electric Power Company (TEPCO) and the Tokai Daini Nuclear Power Station of Japan Atomic Power Company (JAPCO). In TEPCO, the dry storage facility is 72 m length x 30 m width x 20 m height, and five larger metal casks, with a capacity of 52 BWR-assemblies per cask, and four smaller metal casks, with a capacity of 37 BWR-assemblies per cask, were introduced in 1995. The facility has a capacity of 20 casks and 150 t HM. These casks are being installed horizontally in the building.

In JAPCO, a new dry storage facility is 54 m length x 26 m width x 21 m height, and four metal casks, with a capacity of 61-BWR-assemblies per cask were introduced in 2002. The capacity of the facility is 24 metal casks, or 260 t HM and 61 BWR assemblies can be loaded into each cask. These casks are stored vertically in the building.

The self-inspection on the cask was carried out by TEPCO. The objectives of self-inspection are to confirm the containment of the metal gasket and the spent fuel integrity. The self-inspection was carried out on casks in 2000 and 2005 after five and ten years of dry storage, respectively. As for the containment, visual inspection on the seal surface at the primary lid and gasket showed no abnormal features. For spent fuel integrity, any ^{85}Kr gas was not detected by gas sampling in cask cavity, and visual inspection on the fuel assembly surface did not show abnormal features.

(6) Spain

Dual-purpose (storage and transport) metal casks have been designed, licensed and manufactured to store spent fuel from the Trillo Nuclear Power Plant. Each cask can store, in a helium environment, 21 PWR fuel assemblies with a maximum burnup of 40 GWd/t U, 4% initial enrichment and a minimum of 5 years cooling time. The casks are initially licensed for a period of 20 years for the storage mode and for 5 years for transport. The first loading was in 2002, presently there are 80 ENSA-DPT casks in storage.

A new facility for the storage 377 fuel assemblies (20 of them damaged) with relatively low burnup and low initial enrichment from the Jose Cabrera NPP under decommissioning are to be stored in 12 Holtec HI-STORM concrete casks.

(7) United Kingdom

The general experience with dry storage of Magnox fuel at Wylfa Power Station, both in the short term CO_2 cooled stores and in the two longer-term air-filled stores, has been very good, although the 2 air cooled modules no longer hold spent fuel.

The fuel elements are held in skips, which consist of 12 x 16 arrays of blind tubes, open at the top, and cooled by air which passes from below over the outside of the tubes, each of which holds one element. The air pressure is maintained slightly negative with respect to atmospheric pressure via an exhaust fan and a line of filters. This ensures that any leakage is inward, and that all air released to atmosphere is filtered. As fuel elements are added and removed from the store, the skips move along conveyor belts, i.e. the location of the fuel elements with regard to the building walls is not fixed.

The Wylfa Diverse Discharge Route (DDR) has been constructed beneath dry cell 4 to enable export of fuel direct from the DSC4 and 5 into off site transport flasks. The DDR comprises a shielded Flask Filling Cell (FFC) with remote fuel handling manipulator and outside the FFC, a number of flasks operating and handling stations together with a flask export station.

(8) United States

- Status

Most nuclear power plants in the United States were not originally designed with a pool storage capacity sufficient to accommodate the spent fuel generated over the operating life of the power plants. Utilities originally planned for spent fuel to remain in the spent fuel pool for a few years after discharge, and then to be sent to a reprocessing facility. Given that the reprocessing option was abandoned in 1977 and no other option for spent fuel disposition currently exists, utilities have expanded the storage capacity of their spent fuel pools by using high-density storage racks. This has been a short-term solution, however, given that most utilities have reached, or soon will reach, their spent fuel pool maximum storage capacity.

Utilities have developed independent spent fuel storage installations (ISFSIs) as a means of expanding their on-site spent fuel storage capacity on an interim basis until a geologic repository is available to accept spent fuel for permanent storage. By the end of 2007, 41 independent spent fuel storage installations were licensed. There was ~12 600 t HM, or ~18 percent of the total spent fuel inventory of the country in dry storage at commercial and Government storage sites.

- Dry Storage Beyond 20 Years [16]

Virginia Electric Power Company (VEPC) was the first licensee to apply for extending its Surry ISFSI license beyond twenty years. VEPCO, by then Dominion, submitted a license extension application in 2002. The original approach was for VEPCO to request a 20-year license extension, but in a separate submittal, VEPCO requested consideration for a 40-year license extension. The NRC began its review in 2003, and following a round of request for additional information in late 2003, issued, in February 2005, a favourable safety evaluation report granting a 40-year extension. The technical basis documenting (1) the absence of aging mechanisms that would threaten the integrity of spent fuel in dry storage, and (2) the excellent performance of the storage system components was based on the results obtained in a joint NRC-EPRI-DOE project that consisted in re-opening a 15-year old CASTOR V/21 dry storage system located at Idaho National Laboratory, and non-destructively and destructively examining its components and contents.

Later in 2005, Progress Energy, the H.B. Robinson ISFSI licensee, received a favourable safety evaluation report, also for a 40-year license extension. The combination of successful applications by the Surry and H.B. Robinson ISFSIs is notable because they cover the range of dry storage technologies that have been implemented at the U.S. (and most worldwide) reactor sites.

On August 4, 2008, the NRC made available preliminary draft rule language to amend its regulations concerning the initial and renewal license periods for Independent Spent Fuel Storage Installations (ISFSIs) and cask Certificate of Compliances. The present term limit of 20 years is being proposed to be extended to 40 years.

- Dry storage of spent fuel with burnup in excess of 45 GWd/t U [18]

In July 2002, the NRC published Interim Staff Guidance 11 (ISG-11), Revision 2 ‘Cladding Considerations for the Transportation and Storage of Spent Fuel.’ This revision contained guidance and criteria applicable to all commercial spent fuel burnup levels and cladding materials, and enabled the placement in dry *storage* of all commercial fuel licensed by the NRC for power plant operations in a manner consistent with the regulations. A further revision (ISG-11, Rev. 3) was issued in November 2003. This revision modified Rev. 2 (1) by clarifying the meaning of some of the acceptance criteria contained in Rev. 2, and (2) by adding criteria to allow higher temperature limits for low burnup fuels. However, it should be noted that contrary to the mention of ‘transportation’ in the title of ISG-11, Revisions 2 and 3 did not provide criteria for the transportation of high burnup (i.e., >45 GWd/t U) fuel.

A peak cladding temperature of 400°C is specified, unless it can be shown that cladding hoop stress are maintained below 90 MPa. For the latter, a higher peak cladding temperature may be calculated. This could be useful for spent fuel characterized by low to moderate burnups, benign fuel duty cycles, or both. There are also restrictions on thermally cycling the spent fuel when the temperature swings are greater than 65°C.

With publication of ISG-11, Rev. 2/3, dry storage of ‘high-burnup’ fuel is no longer a regulatory issue whose merits are being debated. There are, however, unresolved issues with regard to the impact of dry storage on the transportability of the spent fuel after long periods of storage, in particular, with regard to the impact of potential changes in the distribution of hydrogen in the cladding as a result of the thermal transient that occurs following the transfer of spent fuel from wet to dry storage and the subsequent slow cooling over many years --if not decades-- of dry storage.

5.3. RESEARCH ACTIVITIES RELATED TO DRY STORAGE

Research activities relevant to dry storage have been carried out since the early 60s and have been reported in earlier BEFAST/SPAR programmes and elsewhere.

(1) Canada

- Previous ERB, CEX Experiments (see also IAEA TECDOC-1343)
- Used Fuel Aging Mechanisms modelling
- Temperature monitoring/modelling during dry storage
- Used fuel characteristics at the beginning of dry storage,
- Determination of loads and vibrations envelope during handling and transportation,
- Measurements of potential corrosion of dry storage container,
- Preservation of information about the used fuel for 100+ period.

(2) Germany

Dry storage was from the beginning a necessary element in the back end of the fuel cycle strategy of Germany. Therefore, an extended R&D was performed to define the conditions for safe storage of spent fuel under dry conditions. The detailed results from the different investigations were put together early into performance prediction codes and those codes were validated in a demonstration programme using original spent fuel and representative storage casks.

Presently, dry spent LWR fuel storage is licensed for single fuel assemblies with rod burnup up to 65 GW·d/ t HM. This allows dry spent fuel storage of reloads with batch averages up to 55 GW·d/ t HM. Closing the nuclear fuel cycle by direct disposal of the spent fuel, combined with prior long-term dry storage, requires that the end-of-life conditions (EOL) of the fuel assembly (FA) be considered for assessing performance of the disposal approach. In order to take care of FA with higher burn up, the loading schemes for transport and storage need to be defined in more detail on specific, instead of enveloping, EOL data for better response to all licensing requirements. Optimized cask loading schemes, taking into account the available FA with specific neutron and gamma emissions as well as the results from statistical performance calculations, provide a better usage of all transport and storage casks. Additionally, post-pile creep behaviour of highly irradiated commercial cladding materials needs to be analysed to extent licensing to even higher burnups.

(3) Japan

R&D for dry cask storage has been carried out mainly by the Central Research Institute of Electric Power Industry (CRIEPI) [20-22]. The first R&D program, undertaken between 1987 and 1991, dealt with metal cask storage technology and its results provided reference data for the licensing of dry cask storage technology in Japan. The main subjects studied were:

- fuel cladding temperature tests and analysis considering heat transfer within the cask;
- creep tests of fuel cladding;
- containment tests of the cask lid structure;
- cask drop tests;

- material tests of ductile cast iron;
- building collapse and heavy objects drop onto cask;
- cask toppling by earthquake.

CRIEPI also carried out from 1992 to 1996 an advanced research program on spent fuel storage technology. Major activities of this new program dealt with the evaluation and related tests on dry storage methods suitable for spent high burnup fuel and MOX fuel in terms of safety and economy, which included application of burnup credit and temperature analysis. The activities also included various designs and development projects for advanced dry storage technologies, such as a new type of fuel basket with efficient heat removal and subcriticality, and an advanced concrete cask with improved safety and economy.

A further R&D program for interim storage of 'Recycled Fuel Resources' (i.e. spent fuel) was started in 1997 to 1999. This program deals with verification tests related to concrete modular storage technology such as horizontal concrete silos and concrete casks and also deals with integrity of spent fuel during dry storage. Demonstration program for concrete cask was carried out from 2000 to 2003. Major activities of this program dealt with the demonstrations for determining concrete cask performance including the fabrication of full-size concrete, heat removal test, seismic test and also dealt with tensile test on spent fuel cladding, pellet oxidation examination and development of non-destructive monitoring technique of spent fuel. Metal cask tests were added and continued in 2004 and run until 2008. In this new program, drop test, airplane crash test and long-term sealability test of lid structure have been carried out. SCC evaluation test on canister is also carried out to clarify basic deterioration mechanism. The schedule of demonstration programs is shown in Table 4.

For neutron shielding materials, the Japan Nuclear Energy Safety Organization (JNES) conducted to investigate material property change on epoxy resin, silicon resin and propylene glycol (PG) water which would be applied to metal cask [23].

TABLE 4. SCHEDULE OF DEMONSTRATION PROGRAM FOR CONCRETE CASK IN CRIEPI

Program Item	2000-2003	2004-2008
Concrete Cask Tests	Phase1	Phase2
a. Basic design	━	
b. Fabrication of full-scale concrete cask	━	
c .Demonstration tests -Heat removal test	━	
-MPC drop test	━	
-Seismic test	━ 1/3scale	━ Full-scale
d. SCC evaluation test		━
Metal Cask Tests	Phase 1	Phase 2
a. Drop test without impact limiters		━
b. Airplane crash test		━
c. Long-term sealability test of lid structure	━	━

The Japan Nuclear Energy Safety Organization (JNES) has been planning and performing tests in order to evaluate fuel integrity in interim dry storage [24]. By 2003, thermal creep testing and irradiation hardening recovery testing had been performed using irradiated cladding tubes and the creep equation for boiling water reactor (BWR 50GWd/t U) and pressurized water reactor (PWR 48GWd/t U) fuel cladding had been established. Since 2004, JNES has been performing the tests using irradiated cladding tubes, in which the effects of hydride reorientation will be investigated for up to 55GWd/t U BWR and PWR fuel cladding.

JNES tests were composed of hydride effect evaluation testing, irradiation hardening recovery testing, and creep testing. In each test, both BWR and PWR irradiated cladding tubes were used as test material. In hydride effect evaluation testing, hydride reorientation testing has been performed in order to evaluate the correlation between hydride reorientation behaviour and conditions such as hoop stress, temperature, and cooling rate for BWR and PWR irradiated cladding tubes. Mechanical property testing is also performed as a part of hydride effect evaluation testing, in which mechanical properties of cladding with radial hydride after hydride reorientation testing are evaluated. Irradiation hardening recovery testing is performed in order to evaluate the irradiation hardening recovery behaviour during interim dry storage. As for creep behaviour, JNES had already established the creep equation based mainly on secondary creep rate evaluation for 50GWd/t U BWR and 48GWd/t U PWR fuel cladding tube until 2003. The creep rupture behaviour had been evaluated for the same type of cladding tubes. The creep property of both BWR and PWR 55GWd/t type fuel cladding tubes will be evaluated by 2006. The test schedule is shown in Table 5.

TABLE 5. JNES TEST SCHEDULE

		2000	2001	2002	2003	2004	2005	2006
Survey and planning		▬				▬		
Creep test	Creep test	PWR 48 GWd/t U, BWR 50 GWd/t U ▬	▬	▬	▬		PWR 55 GWd/t U, BWR 55 GWd/t U →	→
	Creep rupture test	PWR 48 GWd/t U, BWR 50 GWd/t U ▬	▬	▬	▬			
Hydride effects evaluation test	Hydride reorientation test Mechanical property test					PWR 48, 55 GWd/t U, BWR 50, 55 GWd/t U →	→	→
Irradiation hardening recovery test		PWR 48 GWd/t U, BWR 50 GWd/t U			330–420°C ▬		<330°C →	→

(4) Spain

A recent R&D program performed on six high burnup (> 50 GWd/t U) fuel rods that had been dry stored in air for more than 25 years, at temperatures not in excess of 175°C, has proven that the fuel cladding has not suffered any degradation under these conditions.

(5) USA

While issues related to dry storage with burnup greater than 45 GWd/t U have largely been resolved, transportation issues have not, at least for spent fuel with discharge burnups greater than 45 GWd/t U [27-34]. A research program sponsored by industry was launched in late 2002 following two NRC-industry meetings held on September 6, 2002 and October 23, 2002. The aim of the research program was to assess the performance of high-burnup spent fuel cladding under normal and accident conditions of transportation, as prescribed by 10 CFR 71, considering the physical characteristics and mechanical properties of cladding at the end of dry storage.

The results of the analytical studies conducted during the past several years indicate that damage to high-burnup spent fuel under prescribed regulatory conditions of dry storage and transportation will not impair its operational management. Dry storage effects on cladding physical and material conditions, including creep-related deformations, have the potential to impact fuel rod performance during transportation. Specifically, cladding resistance to failure under the dynamic loading of transportation accidents depends on fuel-cladding gap size and radial hydride formation, both of which could—to a limited extent—evolve during long-term dry storage. The results of the hypothetical accident analysis indicate that cladding failure would be bi-modal—taking the form of (1) a state of failure initiation at the cladding inside diameter (ID) remaining as part-wall damage, with less than a 2% probability of occurrence and (2) a through-wall failure with a probability of 1E-5. The response analysis under normal conditions of transport shows a large margin against fuel rod failures. The grids and guide tubes, which form the structural elements of the fuel assembly, are predicted to remain structurally competent. As a result, the geometric form of the spent fuel assemblies will not be substantially altered.

An NRC-sponsored experimental program is being conducted at Argonne National Laboratory. However, the program has incurred substantial delays due to the unforeseen unavailability of ANL hot cells necessary to conduct the work.

6. FUEL INTEGRITY

6.1. EXAMPLES OF DAMAGED FUEL

6.1.1. Damaged fuel rods

Figures 1 and 2 show examples of primary and secondary defects arising in PWRs and BWRs. Many primary defects are induced by small metallic debris. Due to rod vibrations, the cladding can be locally wearied off. A similar situation may occur in case of a very low holding force of a spacer grid mesh if the rod interacts with a spacer grid vane or with the spring itself. Secondary degradation can often be recognized by appearance of hydride blister at characteristic axial locations of the fuel rod. Occasionally, severe secondary degradation, such as long cladding splits, may be observed, Fig. 2 [35].

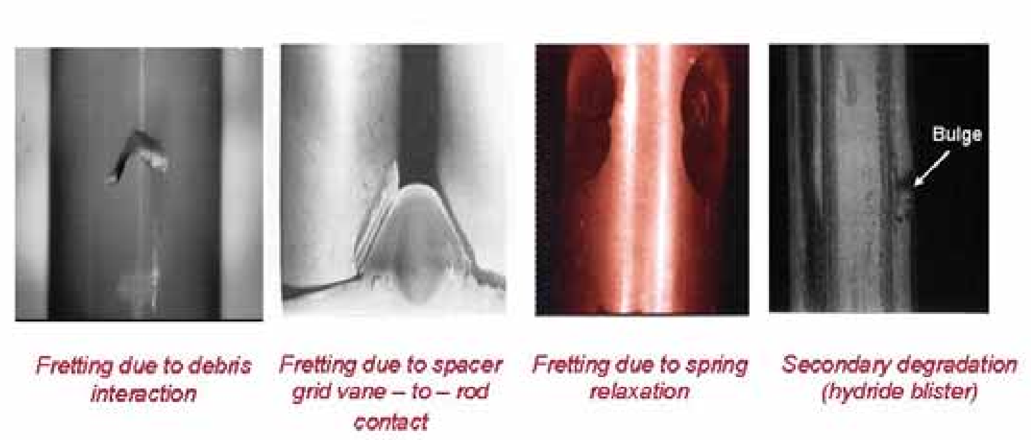


Fig. 1. Fuel rod damages due to fretting and secondary degradation.

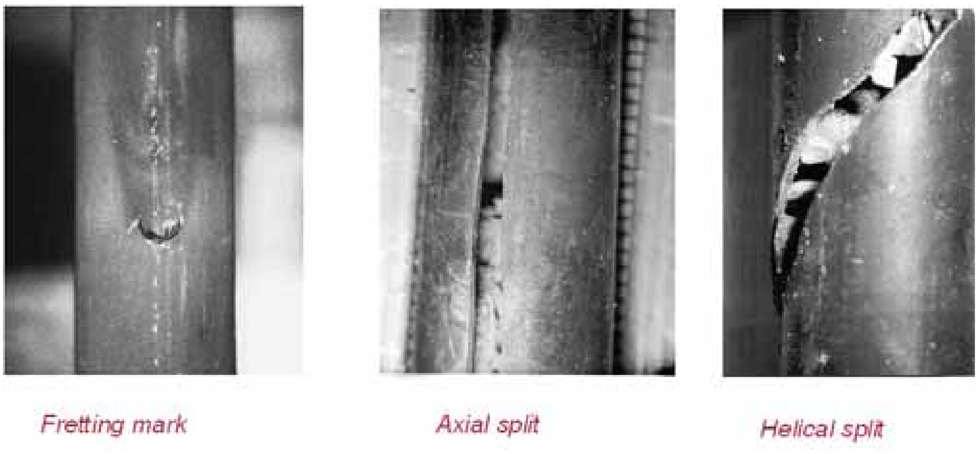
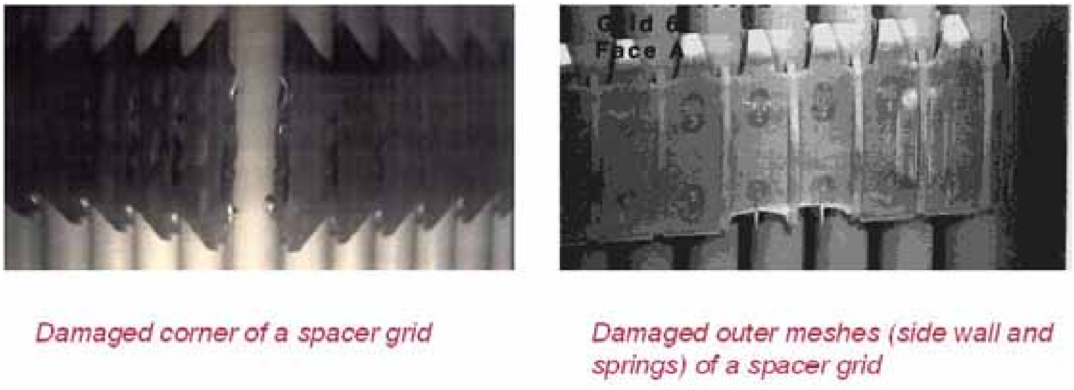


Fig. 2. BWR fuel rod damages.

6.1.2. Damaged structural parts

Structural parts may also show damage. During inspections in the spent fuel pool, great varieties of findings are observed, such as contact and wear marks at the tie plates, fretting marks at spacer grids up to damaged spacer grid corners or meshes as well as broken guide tubes or slightly twisted fuel assemblies. Figure 3 shows on the left side a damaged corner and on the right side damaged outer meshes of a spacer grid. Damages may arise during operation or during handling of fuel assemblies. They do generally not affect fuel rod integrity, but may have an impact on the ability to further handle fuel assemblies in the reactor pool or under storage operations.

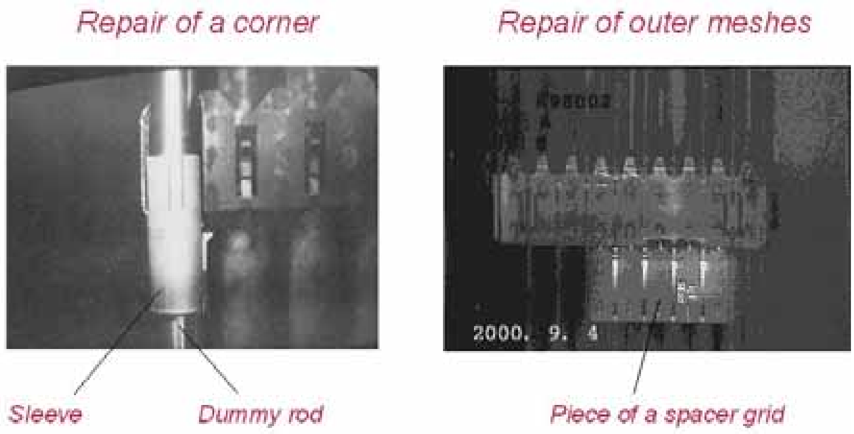

Damaged corner of a spacer grid

Damaged outer meshes (side wall and springs) of a spacer grid

Fig. 3. Examples of damaged spacer grids.

6.1.3. Reconstitution of damaged fuel

Primary objective of a reconstitution of fuel assemblies is to enable their reinsertion for further reactor operation. Main techniques are replacement of damaged fuel rods, repair of spacer grids, and exchange of whole skeletons. Damaged fuel rods can be replaced by new fuel rods or by dummy rods made of stainless steel or Zircaloy. A good deal of techniques is also available to reconstitute structural parts of fuel assemblies for further reactor operation. Some examples are repair of spacer grid corners and outer grid meshes (Fig. 4), recalibration of springs in spacer meshes, adjusting of vanes on spacer grids, exchange of tie plates and hold down springs, etc.


Repair of a corner

Repair of outer meshes

Fig. 4. Repair of a spacer grid corner and outer meshe.

6.2. FUEL INTEGRITY DEFINITION AND CRITERIA

6.2.1. Situation in the SPAR-2 member states

Fuel integrity definitions are originally focused on the fuel to fulfil reactor operational requirements. Given the great variety of reactors and fuel types, it is obvious that terminology and methods used to describe and measure fuel integrity exhibit many nuances all over the world. The following chapters describe the situation in the SPAR-2 member states.

(1) Canada

After irradiation, all used fuel is transferred into wet storage at the reactor sites. Fuel bundles are considered to be defected if the element sheath or endcap has been breached, allowing the release of fission products. Defected fuel bundles are detected during irradiation by fission product monitoring of the coolant. The defected bundles are generally placed in separate baskets in the wet storage bays and are not presently accepted for dry storage. A fraction of discharged fuel bundles are also inspected at discharge for wear and deformation. Fuel with severe wear and deformation is considered damaged and also set aside in the wet storage bays, and not presently accepted for dry storage.

Current plans are for most of the used fuel to be transferred into dry storage until a solution for long-term management is available. Due to the large number of fuel bundles, the transfer must rely on automated processes that are designed for intact bundles. Any abnormalities with the bundles' structural integrity may affect the automated process; for example, loose elements may get stuck in handling equipment. Bundle integrity therefore is important for dry storage, and means that the fuel bundle is structurally sound. This is distinct from bundles that pose a radiological hazard due to the fuel sheath being compromised; this is referred to as element integrity.

(2) France

In France the terminology is agreed between EDF and AREVA NC La Hague.

Failed fuel:

- Gas leaking fuel: fuel assembly not showing visual defects which may jeopardize the containment by the cladding of fissile material, pellets or fragments of pellets.
- Damaged fuel: fuel assembly susceptible to spread fissile material. There is no containment by the cladding of radioactive material, pellets or fragments of pellets.

In France, the number of failed fuels in EDF is 5 to 10 / year, and total amount of failed fuel assemblies at EDF is 0.1%.

Detection of damaged fuel assemblies is done at the NPPs. At fuel discharge, there are sipping tests and visual inspection.

In the past, detected damage was repaired and the fuel assembly was moved back to the reactor, but since 2002, the authority forbids to put failed fuel assemblies back in the reactor. Some defect fuel rods are removed from the fuel assembly, accommodated in a 'quiver' then put in a packaging and transported to La Hague. More damaged fuel assemblies, with broken rods for example, are stored in the pool of the NPP.

(3) Germany

Fuel rods are considered intact if the fuel assemblies are discharged from the reactor without failure indications from the online monitoring system or subsequently by sipping tests. In case that damaged fuel assemblies are detected, the damage is generally repaired to ensure further reactor operation.

A guideline promulgated by the Reactor Safety Commission stipulates that defect fuel rods, which shall be stored in casks, need special measures, such as gas-tight encapsulation or moisture absorbers. The licensing of special capsules that can accommodate defect fuel rods and can be loaded in dry casks is in progress. Zeolite candles are already in use in CASTOR 440 casks loaded with low burnt WWER fuel stored in the ZLN.

During cask drying, sometimes gas leakage of single fuel rods is observed. The defect rate is comparable to the defect rate of reactor operation and, thus, the defects most likely originate from it.

(4) Hungary

Fuel rods are considered intact if the fuel assemblies are discharged from the reactor without failure indications from the online monitoring system or subsequently by sipping tests. In case of damaged fuel assemblies, the damage cannot be repaired owing to the closed (shrouded) construction of the assemblies. Failed fuel should be placed in hermetic bottles, placed in specific positions of the fuel storage rack, and are not permitted to be loaded in the MVDS without additional measures (second barrier). So far no fuel has been identified as failed in the reactor.

(5) Japan

In the regulation in Japan, only intact fuel is allowed for the interim dry storage basically. Here, ‘intact fuel’ means fuel which didn’t show any leakage during service in reactor and any geometry change on the fuel assembly such as fuel rod bowing and position displacement of assembly components. The reasons for non-leakage are based on the idea that it is difficult to evaluate the effect of residual moisture in leaker fuel on inert atmosphere and fuel cladding hydrogen embrittlement caused by pellet oxidation. The reason for no change of geometry on the fuel assembly is that the safety analysis in the regulation authority is carried out based on a right position of fuel assembly components.

On the other hand, clear definition of ‘fuel integrity’ during storage is not described specially.

As for Regulation concerning interim dry storage in Japan, ‘Technical requirements in safety examination for spent fuel interim storage facility using concrete cask’, ‘Technical requirements in safety examination for spent fuel interim storage facility using metal cask’ from NISA (Nuclear and Industrial Safety Agency), and ‘Safety review guideline for interim storage facility’ from NSC (Nuclear Safety Commission of Japan), were published. In each document, storage facility or metal cask is required to be designed to be able to maintain the fuel integrity.

The description in the technical requirements concerning fuel integrity are as follows, ‘Temperature of the spent fuel should be maintained to be less than the restricted value to avoid any creep failure and mechanical property degradation of fuel cladding’, ‘Fuel material properties used in the safety analyses must have appropriate safety margin’.

In the creep failure evaluation, 1% creep strain is adopted as the criteria.

As ‘mechanical property degradation of fuel cladding’, some concrete phenomena, such as radial hydride reorientation and the irradiation hardening recovery, are mentioned as the items to be considered. The concrete criteria for these phenomena are now under preparation considering the test results.

As for ‘fuel material properties used in the safety analyses’, material property change during both services in reactor and storage, such as the effects of absorbed hydrogen and the effects of fast neutron irradiation, are required to be considered.

(6) Slovakia

The fuel assemblies are controlled by sipping tests. Leaking fuel is placed into special bottles and transported to the Bohunice interim storage pools. Presently, a sophisticated manipulator is prepared for dismantling and disassembling operations for these assemblies. After acceptance testing of this apparatus, new criteria will be set in 2008, how to manage damaged fuel in future.

(7) Spain

A working group formed by the utilities, the regulator, the main fuel vendor and the national radwaste agency has been created with the aim to develop a guide to define damaged fuel for the purposes of storage and transportation and to provide practical guidelines for its implementation.

Current draft defines damaged fuel in this context as a fuel assembly for which the rod integrity or the assembly geometry cannot be guaranteed. As such, specific measures must be taken to manage these assemblies in order to provide:

- confinement for particulates which might escape from the rod,
- means for handling and retrievability,
- confinement of the assembly in a known volume in order to guarantee the assumptions taken in the confinement, shielding and criticality analyses.

(8) Sweden

Fuel rods are considered intact if the fuel assemblies are discharged from the reactor without failure indications from the online monitoring system or subsequently by sipping tests.

If leaking spent nuclear fuel is transported from a NPP to the interim storage CLAB, fuel with Cs-137 concentration which increase by more than 5 E4 Bq/kg the fuel is considered as leaking.

(9) UK

The following definitions are used to categorise spent oxide fuel (for reprocessing) in accordance with Oxide Fuel Plant Safety Cases at Sellafield, UK.

For LWR fuel, the following definitions are used.

- Standard fuel – fuel that complies with Plant Safety Cases as follows:
  - $\leq 4\%$ ^{235}U enrichment, ≤ 40 GWd/t U *
  - structure 'as manufactured';

* Note: Fuels with higher enrichment and irradiation have been received and reprocessed.

- Failed (leaker) – fuel that has a breach of cladding integrity, e.g. minor holes, cracks or end cap failure resulting in release of mobile fission products only.

- Non-standard – fuel that is out with Reference Case;

examples are fuel assemblies with

- non-standard dimensions
- reactor channels still in place
- missing pins or additional pins
- reconstituted structures/components
- non-standard additives, e.g. Nb, Cr

- Damaged – fuel which has undergone a geometric change to the 'as manufactured' structure which would require a safety case revision or cause fuel handling changes.

examples are fuel assemblies with:

- deformed pins
- damaged or missing top nozzle
- fractured tie rods
- additional support to ensure integrity (wires, bars)

For AGR fuel, the following definitions are used.

Only covers the receipt of standard fuel and failed / damaged fuel that has been conditioned

- Standard
  - bounded by individual fuel storage plant Safety Cases
  - min 60 days cooled
  - max irradiation 40 GWd/t U
  - max enrichment 3.8% ^{235}U
  - structure – as manufactured (for dismantling at Sellafield)

- Failed - pins with breached cladding: up to 7 pins, whole or in cut pieces, packed in capsules (see section 1.3)

Fuel that has been handled through hot cell facilities and subsequently placed in PIE cans, examples are:

- intact fuel pin residues from dismantled fuel elements
- punctured fuel pins
- intact fuel pins from non-destructive analysis
- fuel fragments and U_3O_8 powder from destructive analysis.

(10) USA

In the USA, ‘damaged’ fuel is defined as fuel with cladding defects greater than hairline cracks or pinholes, (at least in the regulatory context applicable to spent fuel storage and transportation.) NRC Interim Staff Guidance 1 [36] provides qualitative guidance with regard to the classification, and subsequent handling, of spent fuel in three categories: intact (i.e., no defects or defects no greater than hairline cracks or pinholes), damaged (i.e., containing defects greater than hairline cracks or pinholes), and debris (such as fuel rod segments).

ANSI Standard N14.33-2005 [37] defines terms related to dry storage and transport of damaged light water reactor (LWR) spent nuclear fuel. It establishes procedures for identifying, categorizing, and managing damaged fuel. The standard provides:

- Methods for identifying and classifying damaged and undamaged spent nuclear fuel assemblies;
- Preparation and handling requirements for damaged spent nuclear fuel assemblies for dry storage and transport;
- Requirements for record keeping and quality assurance; and
- Specifies the requirements for canning damaged fuel assemblies.

6.3. FAILURE CLASSIFICATION

In general, fuel integrity is strongly correlated with the measurement of activity coming from leaking fuel. This means, the criteria for damaged fuel is primarily based on online monitoring and sipping systems, respectively, which detect rod cladding failures, only. Mechanical aspects are usually considered on a case-by-case basis.

As a result of this situation, current integrity criteria for interim storage tend to refer to this kind of damage definition.

6.3.1. Determination of spent fuel integrity

Typical techniques to monitor defective fuel are online monitoring, sipping and leaching tests, and detailed examinations on fuel assemblies, such as visual inspection, ultrasonic examination, or eddy current testing.

All reactors have at least one on-line monitoring system for the detection of in-core fuel failures as a means of controlling the contamination of primary cooling circuits and subsequent dose to personnel during maintenance operations. In LWR the onset of a fuel failure is usually detected by monitoring the gamma-activity level associated with specific radioactive fission products in the reactor coolant or off-gas. A typical failure indication is an increase in Xe, Kr or I isotope activity. However, to characterize the failure it has to be distinguished between fission products coming from tramp uranium or defective rods. In CANDU reactors, the delayed neutron (DN) system measures the activity that comes from ^{137}I and ^{87}Br (they release neutrons and are also β and γ emitters. An increase of the DN signal indicates uranium release to the coolant [38].

In addition to the in-core systems, most reactors also have secondary sipping systems associated with defuelling and in-pool operations to check cladding/fuel assembly integrity. Fuel failure detection by sipping is based on the release of gaseous fission products from the leaking rod. The samples can be taken either from the top of the fuel assembly or the fuel assembly can be inserted in a closed can. Driving forces for the release of the fission products may be heating up due to reduced water cooling or pressure decrease due to lifting of the fuel assembly.

In PWRs the sipping is done during unloading, it requires only about one additional minute per assembly and is done routinely, since the cores are always unloaded. In BWRs partial sipping is common, especially in combination with a previous flux-tilt to define a narrow area in the core where the leaking fuel is. For WWER special sipping systems were supplied. Direct samples can be taken from the fuel in the opened reactor core by a sampling bell. The fuel assemblies can also be sipped in a can.

The oldest system is Can Sipping, which goes back into the 1970s, Fig. 5.

Today, advanced mobile systems for both PWR and BWR fuel assemblies are in use, see Fig. 6. For cold fuel, additional electrical heater elements are implemented in the can.

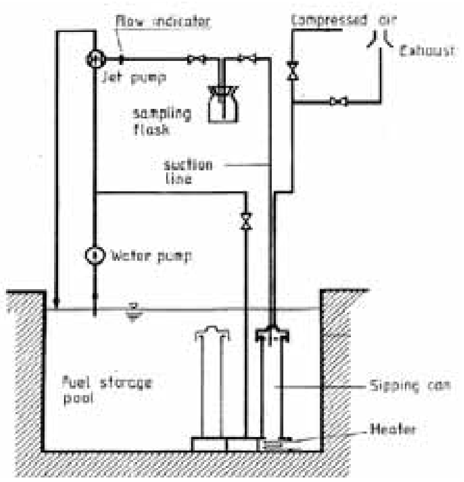


Fig. 5. Flow diagram of a can sipping system.

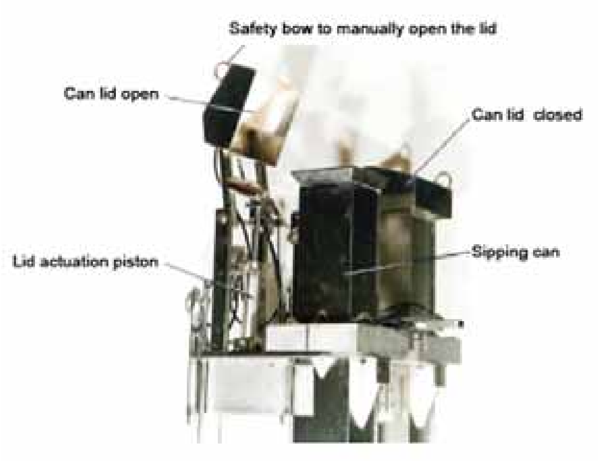


Fig. 6. Upper part of a transportable can sipping system.

Mast Sipping is performed by means of a suction system attached to the fuel assembly manipulator mast. Therefore, it can be carried out during reactor unloading or any fuel assembly shuffling operation. Fuel assemblies, which are confined in water channels and thus separated from one another, can be tested by sipping hoods placed on top of the reactor core covering an array of up to 16 fuel assemblies, Fig. 7 and Fig. 8.

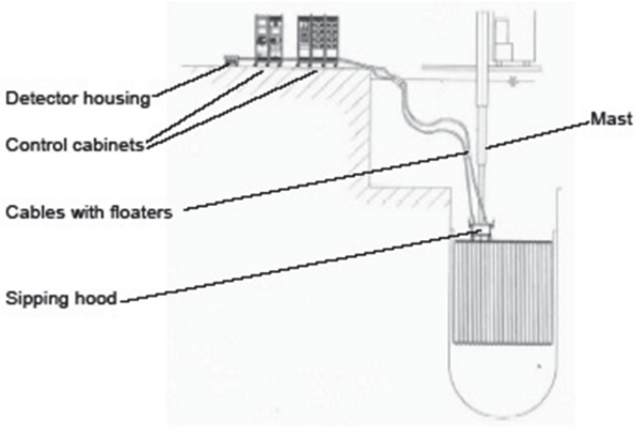


Fig. 7. Positioning of the sipping hood in a BWR.

Fig. 8. Sipping hood for 16 fuel assemblies.

In France, in order to find the damaged fuels, first sipping test is performed at reactor (first sipping) when the fuel assemblies are discharged.. Sipping method is based upon analysis of ^{133}Xe, which can be measured in the pool water after lifting of the fuel assembly or by using an air flow which picks up the fission gas when the fuel assembly is positioned at a high water level in the pool, Fig. 9. From this inspection, fuel assemblies are classified as intact, damaged, or suspect.

In case of 'suspect', a sipping test with a second method is carried out in a cell of the fuel building (Fig. 10.): Thanks to the heating in a bottle ($\Delta T > 30°C$) there is an increase of rod internal pressure. Analysis of ^{133}Xe or ^{85}Kr: conclusion for the fuel may be 'intact' or 'suspect', and if it is suspect a new analysis of ^{134}Cs and ^{137}Cs is done, to conclude either 'intact' or 'failed'. This second sipping test method is very reliable (99%).

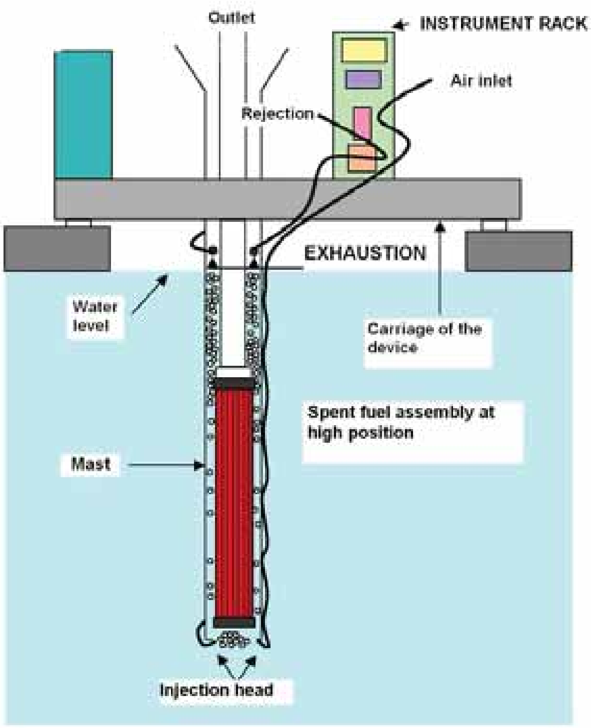


Fig. 9. First sipping at reactor (air flow assisted method).

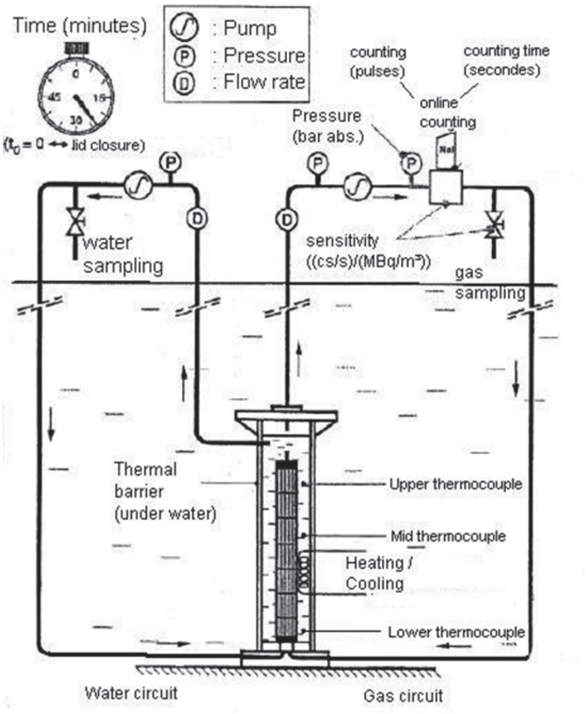


Fig. 10. Second sipping in case of suspect fuel.

In the reactor pool, further techniques can be applied to characterize the integrity of the fuel assembly, such as visual inspection, endoscopy, ultra sonic and eddy current defect testing. Whereas sipping is performed on at least a whole fuel assembly, these techniques are geared to identify defective fuel rods or local damage of the structure which may need to withdraw a considerable number of fuel rods.

An example for damaged fuel rod detection by ultrasonic method (ECHO330 developed by AREVA NP, Fig. 11,) is shown next, while the detection principle is shown in Fig. 12 [39].

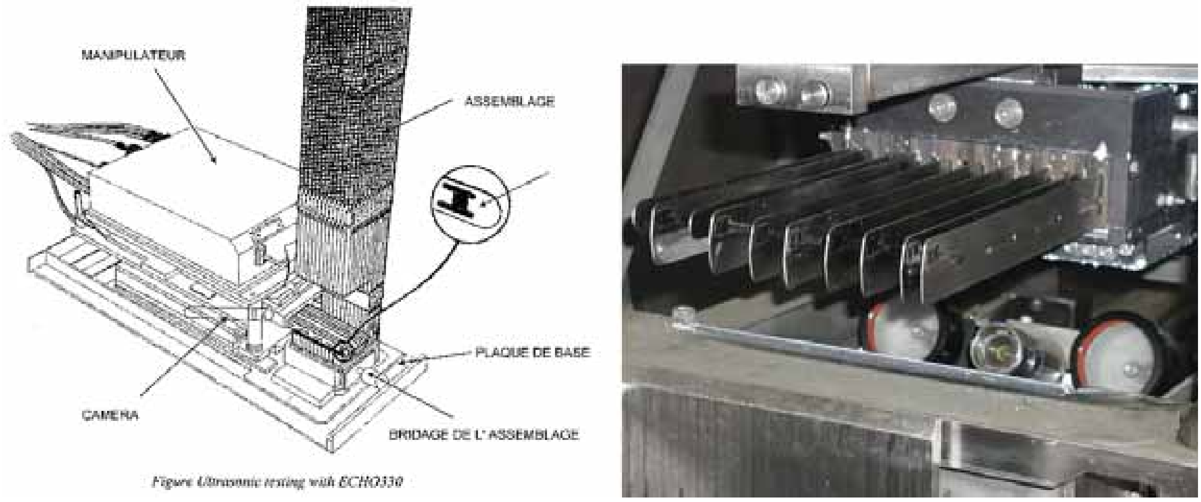


Fig. 11. Ultrasonic testing with ECHO330.

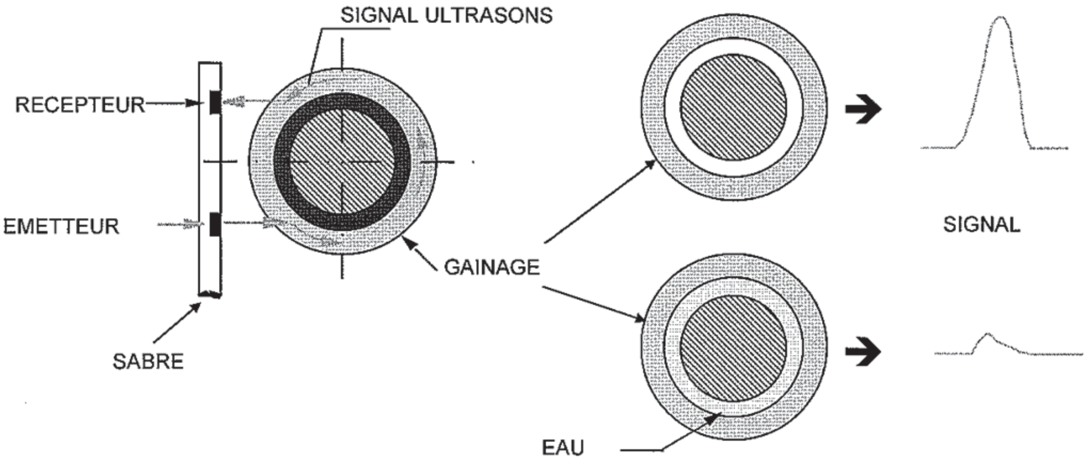


Fig. 12. Principle of failed rod detection by ultrasonic.

The method is reliable, but since it is developed for damaged rods upon the presence of water between cladding and pellet, there are 2 difficulties:

- There is sometimes no water in the lower part (underestimation of failed rods)
- Pellet cladding interaction (overestimation of failed rods)

6.4. DAMAGED FUEL (PACKAGING AND RETRIEVAL)

6.4.1. Encapsulation technique for damaged fuel rods

A new generation of gas-tight capsules was developed by AREVA NP that uses loosely screwed end plugs to drain the capsule by pushing away the water with inert gas, Fig. 13. The built-in filter retains particles inside the capsule. After draining, the capsule will be finally closed. All activities can be carried out in the spent fuel pool in the course of standard nuclear fuel services. The gas-tight capsules can be loaded in canisters having dimensions of PWR or BWR fuel assemblies, Fig. 14. The drained and gas-tight fuel rod capsules and capsule canisters are designed to meet all requirements in terms of pressure, temperature and leak tightness arising in connection with at-reactor and away-from-reactor spent fuel storage, reprocessing and shipment together with irradiated fuel assemblies.

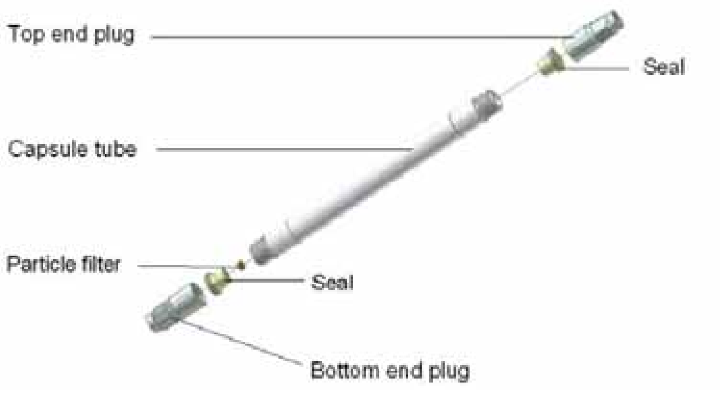


Fig. 13. Fuel rod capsule (gas-tight and water drainable).

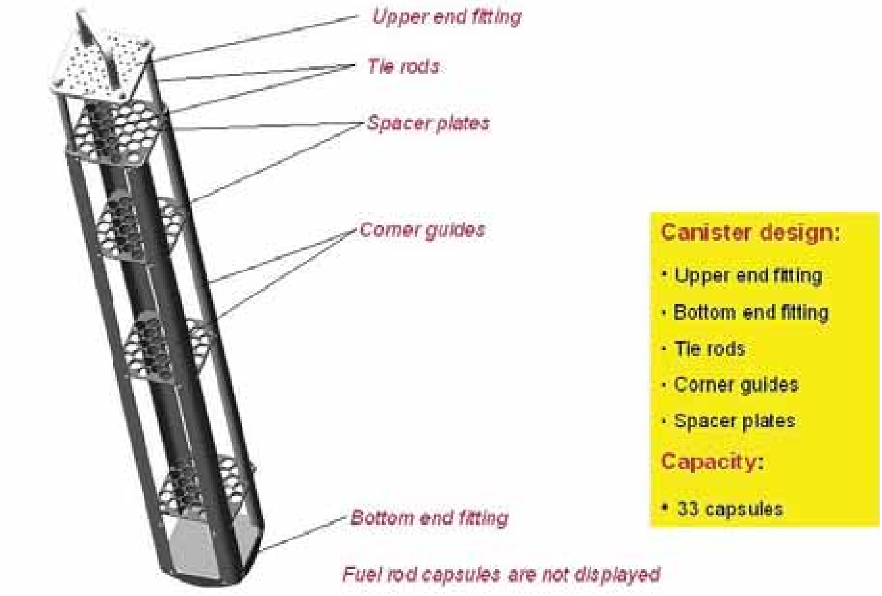


Fig. 14. BWR canister for fuel rod capsules.

6.4.2. Handling of damaged WWER fuel at the ZLN

After the final shutdown of the nuclear power plants in Greifswald and Rheinsberg 3 significantly damaged WWER fuel assemblies had to be loaded into CASTOR-casks of type KRB-MOX. To this end, the different parts of the damaged fuel assemblies were packed by special gripping devices and sleeves in open canisters placed in the cooling pond. New baskets were constructed to accommodate the canisters and successfully loaded into the CASTOR KRB-MOX cask, Fig. 15. Due to low heating power of the fuel, the cask had to be heated from outside to remove water.

Fig. 15. View on the top of the loaded CASTOR KRB-MOX showing basket and primary and secondary lid flange.

6.4.3. Handling and storage of fuel debris at Paks NPP

On 10 April 2003, during the outage period, a chemical cleaning program for the fuel assemblies has been carried out at the unit 2 of Paks NPP, in a specially designed cleaning tank. Thirty fuel assemblies have been significantly damaged due to inadequate cooling. The recovery from this event took about 3 years.

The damaged fuel was collected and taken out manually from the cleaning tank using small cranes, hand operated manipulators and other instruments. The layout of the equipment is shown in Fig. 16. Fuel debris and larger fuel assembly fragments were placed into specially constructed continuously vented canisters. The size of these vessels was designed to take into account subcriticality considerations for bulk fuel material.

Fuel structure materials without fissionable material content were placed in full size canisters. All canister dimensions were such that they fitted the existing spent fuel pool racks and the coupling of the refuelling machine and thus could be located in the spent fuel storage rack positions for temporary storage (5 years).

During this period the canister is vented (passing the bubbles of the radiolysis gases through a gas lock and keeping the radioactive water inside the canister by a lengthy helical tube mounted in compensators on the top of the canisters). Upon expiry of the interim storage, water will be drained, the canister will be dried out, and new metallic O ring hermetic sealing will be installed with catalytic recombiners attached to the canister head. It is expected that the canisters will be shipped for reprocessing.

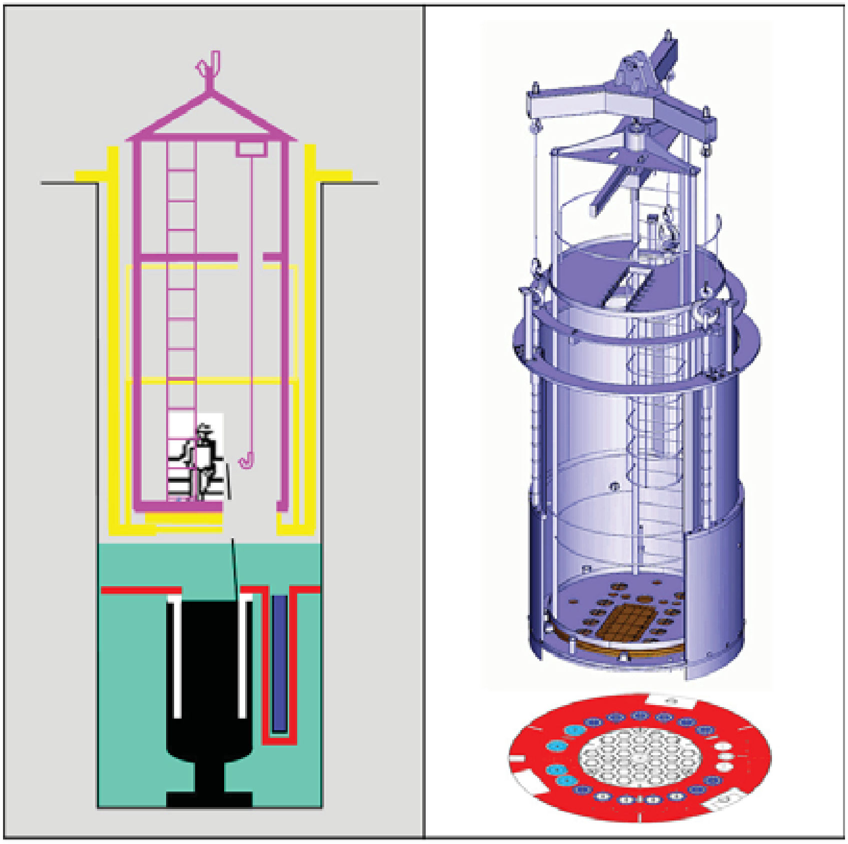


Fig. 16. Layout of the underwater and dry parts of the equipment, 3D image of the fuel management platform and separating rim.

6.5. TRANSPORT OF DAMAGED FUEL ASSEMBLIES

(1) France

For transport, damaged (gas leaking) fuel assemblies in France are transported in bottles accommodated in the transport packaging. At La Hague, only intact or gas leaking fuel assemblies are accepted. Fuel assemblies susceptible to spread fissile material are not accepted.

The bottle is an internal canister which provides a relative containment (Fig. 17.). A new package design for transporting damaged (gas leaking) fuels without using bottle has been applied for license in France recently.

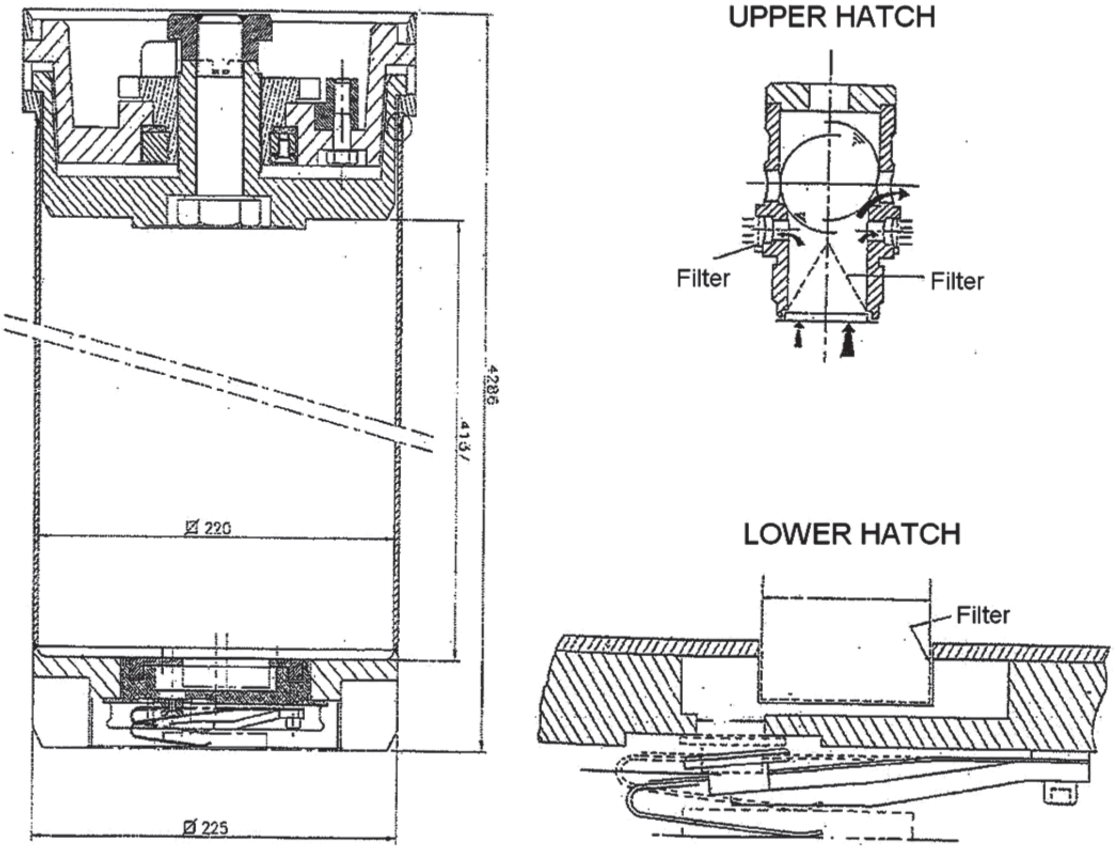


Fig. 17. Bottle for damaged (gas leaking) fuel assemblies.

Before unloading the fuel assembly at La Hague Reprocessing Plant, different inspections are performed.

Control of the atmosphere of the cavity: pressure and activity (^{85}Kr)
Cooling Water activity (^{137}Cs)

The results are satisfactory: no new defect coming from transport has been found.

Then, failed fuels are reprocessed in the same way as intact fuels.

(2) Sweden

Transport of minor leaking fuel from NPPs to CLAB has been executed. The experience is good and no further damage to the fuel because of the transport and cooling process at the reception at CLAB was observed. Prior the transport the fuel assembly is inserted into a special box that is open in the bottom and in the top by valves that also contain filters. The purpose of the box is to prevent any possible fuel fragments/debris that can come loose from the fuel to contaminate the transport cask and the cooling system at CLAB. When the fuel has been unloaded at CLAB the fuel assembly will be taken out of the box and it will be treated as an intact fuel assembly under the condition that the fuel does not have any major damage as broken rods or similar damage.

7. FUEL ASSEMBLY DEGRADATION MECHANISMS IN DRY AND WET STORAGE

A general description of several types of fuel assemblies, or fuel bundles, is given in Section 7.1. The significant degradation mechanisms that may affect the integrity of spent fuel assembly/bundle structure during interim storage, with particular emphasis on fuel rod cladding, are reviewed in Section 7.2. The potential effects of transferring spent fuel from wet to dry storage, or from dry to wet storage, are briefly described in Section 7.3.

7.1. MAIN FUEL ASSEMBLY COMPONENTS

Maintaining fuel assembly structural integrity is essential to facilitate safe handling of spent nuclear fuel. There are two important factors to consider:

- Maintenance of assembly/bundle structural integrity in order to avoid operational difficulties during handling.

- Maintenance of fuel bundle geometry to minimize variations in the fuel’s nuclear reactivity.

7.1.1. LWR

LWR fuel assemblies comprise fuel rods and numerous structural components. Some modern fuel assemblies used in the European market are displayed in Fig. 18.

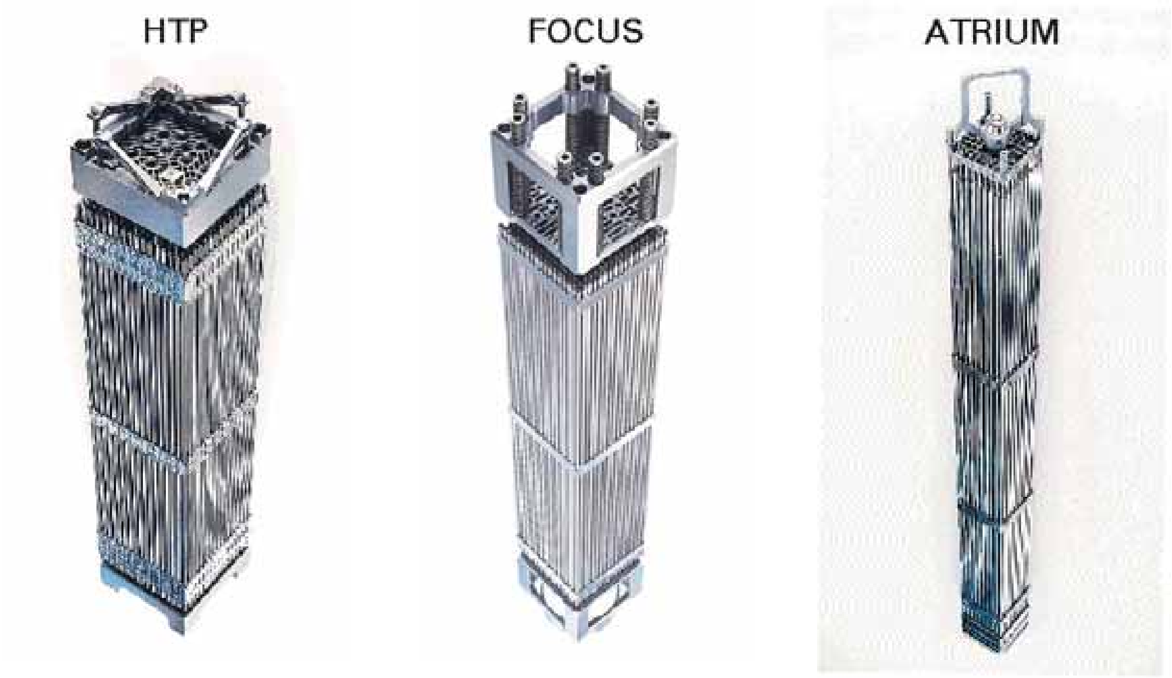


Fig. 18. Fuel Assemblies for PWR (HTP and FOCUS) and BWR (ATRIUM).

All PWR and modern BWR fuel assemblies/bundles contain fuel rods equally spaced from each other. Cylindrical uranium oxide pellets are inserted in zirconium-based alloy tubing. Western PWR assembly designs consist of fuel rods bundled in a 14x14 to 18x18 square array; Russian designs are hexagonal and ‘canned’, i.e., they are surrounded by a thin shroud. BWR designs consist of rods bundled in a 7x7 to 10x10 arrays, and are also canned. The different structural components of the fuel assembly, such as spacers, guide tubes, top and bottom end nozzles, tie plates and fuel channels are manufactured mainly from zirconium-based alloys, stainless steel, and Inconel.

Wet storage performance of LWR fuel structures has, in general, been excellent. In a few cases, stress corrosion cracking in bulge joints connecting the stainless steel grid sleeves to the Zircaloy guide thimbles has been reported in PWR fuel assemblies. This corrosion phenomenon is related to sensitized 304 stainless steel in the presence of adverse pool chemistry conditions, such as the presence of sulphate or chloride ions. This has negatively impacted spent fuel assembly handling [40].

In dry storage of the spent LWR fuel, no degradation mechanisms of the fuel assembly structure are known to be of concern and no detrimental experience has been reported.

7.1.2. AGR

The main components of the AGR fuel assembly comprise 36 fuel pins held in a graphite sleeve by a bottom grid plate and two braces along the length of the assembly (Fig. 19). Except for the outer graphite sleeve, all other components are made from highly alloyed stainless steel. In-reactor, up to eight AGR fuel assemblies are joined together by a stainless steel tie bar passed through the centre of each assembly to form a fuel stringer ~8 m in length.

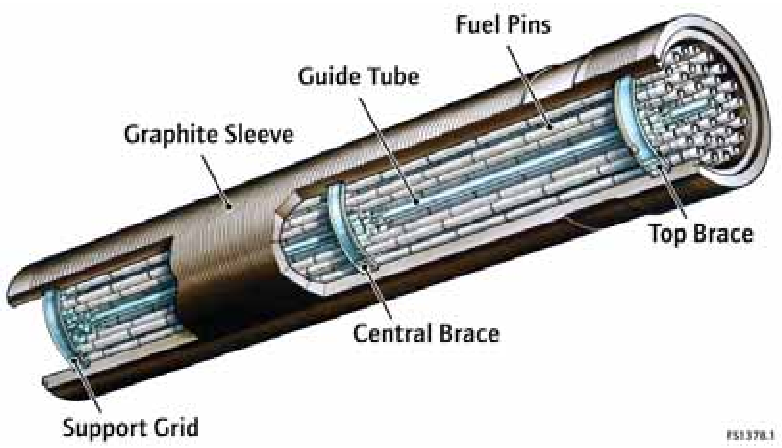


Fig. 19. AGR fuel assembly.

In spite of the AGR being a gas-cooled reactor, the spent fuel is stored wet. The precautions used to ensure a satisfactory fuel rod performance in wet storage also ensure the stability of the AGR fuel element structure.

7.1.3. Magnox

The designs of Magnox fuel elements, although similar, are tailored for each reactor. Each element basically comprises an inner natural uranium rod machined with groves to reflect its core position, and one of either two types of outer hollow extruded and machined magnesium alloy can (polygonal or 'herringbone'); a cone end fitting, ceramic insulating discs and machined end cap complete the element. The inter-space is charged with a small amount of helium prior to the outer can being locked into the groves in the uranium rod by externally pressurizing the sealed element. In the reactor, the elements are placed on top of one another forming stringers within the graphite channel. The purpose of machining groves into the natural uranium rods and locking the external cans into them is to minimize localized stressing of the cladding during irradiation (See Fig. 20). The end cap is the only structural component of Magnox fuel.

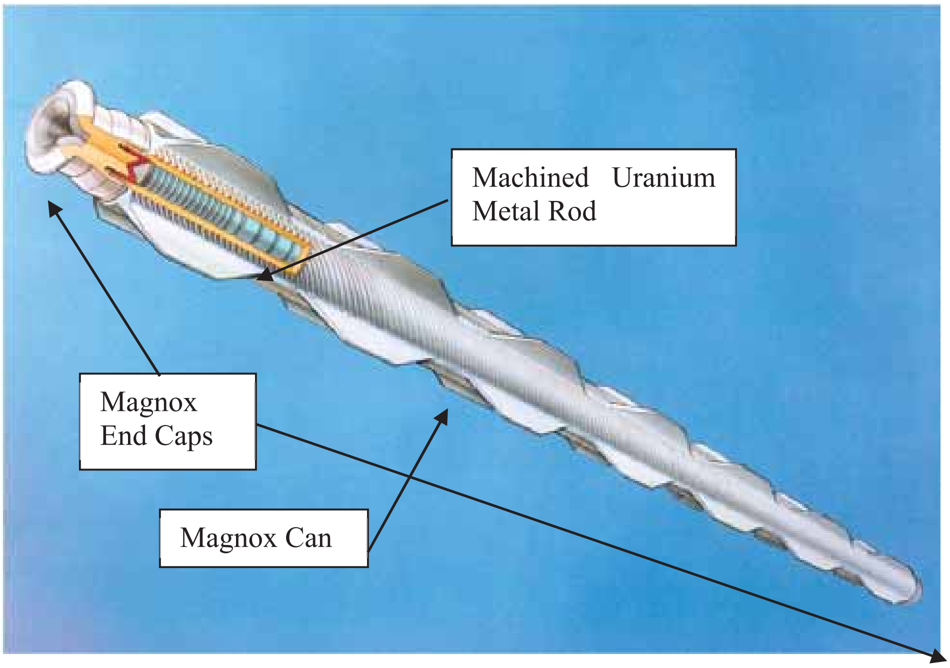


Fig. 20. Magnox fuel.

7.1.4. CANDU

The fuel bundles used in modern CANDU reactors are composed of either 28 or 37 fuel rods, held together in a cylindrical array by welds that attach the end caps of each fuel rod to two Zircaloy endplates (Fig. 21). Each fuel rod consists of a Zircaloy-4 tube containing a string of UO_2 pellets. The overall length of the fuel bundles is approximately 50 cm. Depending upon the location of the fuel rods within the bundle assembly, either spacers or bearing pads are brazed to the outside surface of the cladding. The spacers are designed to maintain coolant flow around the rods while the bearing pads protect the cladding from wear due to friction with the pressure tube during reactor operation. Zr-5Be brazements are used for attaching the Zircaloy spacers and bearing pads to the Zircaloy fuel sheath.

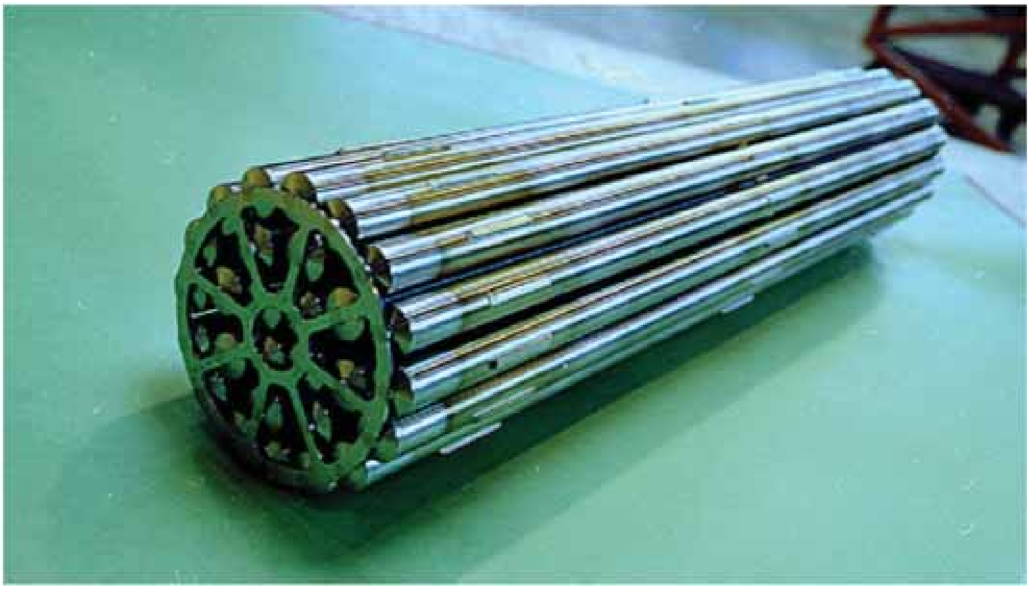


Fig. 21. CANDU fuel bundle (~50 cm in length).

7.2. DEGRADATION MECHANISMS AFFECTING FUEL CLADDING

Maintaining fuel cladding integrity is important because the cladding acts as the primary containment barrier for both fuel pellets and fission products over the storage period and during subsequent fuel handling operations. This is valid for ‘unfailed’ fuel assemblies/bundles used in all the types of power reactors considered: LWR, AGR, Magnox and CANDU. For CANDU and some BWR fuel designs, cladding integrity may also be important for fuel bundle integrity, because some of the fuel pins also act as structural elements for the whole fuel bundle.

7.2.1. LWR

Apart from a limited number of early fuel designs, all current LWR fuel rod claddings are made of zirconium-based alloys. In BWRs, Zircaloy-2 is used, whereas in PWRs, Zircaloy-4 and Zr-Nb alloys are the preferred materials.

7.2.1.1 Wet storage

The degradation mechanisms that may affect cladding integrity during wet storage are as follows:

- Uniform (aqueous) corrosion.
- Pitting, galvanic, and microbiologically-influenced corrosion.
- Hydriding.

(1) Uniform Corrosion

The corrosion behaviour of zirconium alloys in pressurised water and water vapour is well known for temperatures above 200°C. Because the corrosion rate follows the Arrhenius law in the linear, post transition corrosion regime, corrosion rates in the pool water at 30°C to 45°C are a factor of $\sim 10^{10}$ lower[2] than those under operational reactor conditions. Therefore, zirconium-based alloys are virtually immune to uniform corrosion during wet storage, at least under controlled chemistry water conditions.

(2) Pitting, galvanic, and microbiologically-influenced corrosion (MIC)

Zirconium alloys are near the noble end of the galvanic series in contrast to the aluminium alloys, which are near the active end of the series. Electrochemical contact between Zircaloy and aluminium would generate enough potential to oxidise aluminium and hydride Zircaloy. The effects of electrochemical interactions between Zircaloy and aluminium, however, are mitigated by the passivation effect of the oxide layer on Zircaloy surfaces, which acts as a barrier to hydrogen uptake. Therefore, for spent fuel, direct contact between bare Zircaloy and aluminium surfaces does not occur in the pool. Passivated stainless steels are also near the noble end of the galvanic series. Therefore, galvanic corrosion has never been observed when direct contact between Zircaloy and stainless steel occurs in nuclear fuels, either in-reactor or in wet storage. Zirconium-based alloys are not susceptible to pitting corrosion under water chemistry conditions existing in spent fuel pools. MIC of zirconium-based alloys has not been observed.

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[2] Putting this in context, the corrosion resulting from one full power day is equivalent to the corrosion that would occur over a period of several millions years in wet storage.

(3) Hydriding

The average hydrogen concentration in zirconium alloy claddings at the end of in-reactor operation is typically between 100 and 600 ppm, depending on material and in-reactor fuel cycle duty. When the fuel is cooled to the storage pool temperature, most of the hydrogen precipitates in the form of hydride platelets, except possibly for the hydrogen that is trapped by irradiation defects. Due to the heat transfer characteristics of water, hydrogen redistribution by thermal diffusion and formation of so-called pits by an Oswald ripening can be ruled out during long-term wet storage at the pool temperatures of practical interest.

7.2.1.2 Dry storage

Potential degradation mechanisms that may affect cladding integrity of LWR fuels during dry storage and subsequent handling and transportation operations are:

- Air Oxidation
- Thermal Creep
- Stress Corrosion Cracking (SCC)
- Delayed Hydrogen Cracking (DHC)
- Hydride Re-orientation
- Hydrogen Migration and Re-distribution

The first four mechanisms have the potential to result in through-wall cladding defects, while the other ones are unlikely to result in failures; rather, they have the potential to impair the ability of the cladding to effectively withstand potentially adverse mechanical challenges resulting from handling or transportation accidents.

A graphic presentation of these effects is shown schematically in Fig. 22 [41].

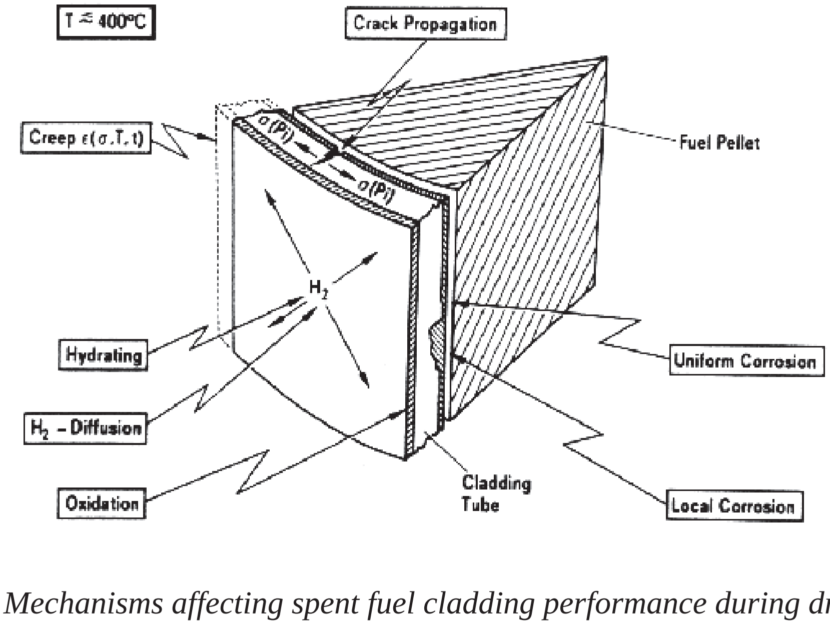

Fig. 22. Mechanisms affecting spent fuel cladding performance during dry storage.

Inert atmosphere in contact with spent fuel during dry storage has been implemented to protect against oxidation. Although creep rupture would most likely lead to a pinhole type of rupture, potential systemic conditions leading to loss of cladding integrity are either prohibited by regulations, or considered undesirable complications even when ruptures can be characterized as ‘pinholes’. To safeguard against the possibility of creep rupture during dry storage, highly conservative criteria were initially established for the amount of creep strain that can develop over the licensed storage period. A 1% end-of-storage-life creep strain was first proposed by Spilker et al. [42] as a sufficiently conservative limit that would be acceptable to regulatory authorities. Significant progress in the state-of-the-art of spent fuel behaviour evolution has been reported during the past five years. As a result, there presently exists a wide variation in regulatory guidance related to thermal creep. The latter covers the field from an adherence to the 1% creep strain limit, as proposed by Spilker at one extreme, to the U.S. situation at another extreme, where no imposition of a restrictive creep strain limit is applied for licensing dry storage systems. Stress corrosion cracking and delayed hydrogen cracking are generally not considered to be active mechanisms for fuel elements with thin-walled cladding. However, DHC is considered to be a limiting degradation mechanism for CANDU bundles.

Much experimental work has been performed to get a better understanding of the conditions under which re-orientation can occur. Motivation for the work is for obtaining a better understanding of the potential effects of dry storage on cladding integrity during subsequent handling and transportation operations. Hydrogen migration due to axial temperature profiles has been found to be of little consequences.

(1) Air Oxidation

Inert atmosphere conditions during dry storage –preceded by appropriate drying and inerting operations– rule out the presence of significant amounts of air in contact with stored spent fuel. Normal conditions do not result in oxidation beyond that incurred during reactor duty. Therefore, oxidation is not an active degradation mechanism under normal inert storage conditions. Air oxidation is possible for off-normal or accident conditions resulting in the failure of the storage system seals. Two scenarios can be considered: (1) fuel rods with no through-wall defects; and (2) fuel rods with through-wall defects.

- Cladding Oxidation

Because oxidation rates of zirconium-based cladding by air are relatively low at temperatures typical of dry storage conditions,[3] losses in cladding wall thickness are limited to a few percents, even for prolonged air exposure. Therefore, cladding remains an effective containment barrier protecting the fuel pellets from exposure to air for fuel rods that do not contain through-wall defects.

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[3] Using the equation developed in *The Corrosion of Zircaloy-Clad Fuel Assemblies in a Geologic Repository Environment* [Hillner E., D.G. Franklin, and J.D. Smee, WAPD-T3173. 1998], it is predicted that only 20 µm of additional cladding would be lost for fuel held at 400°C for 100 days. The Hillner et al.’s equation was developed for steam corrosion, but the corrosion rates in steam and air are similar.

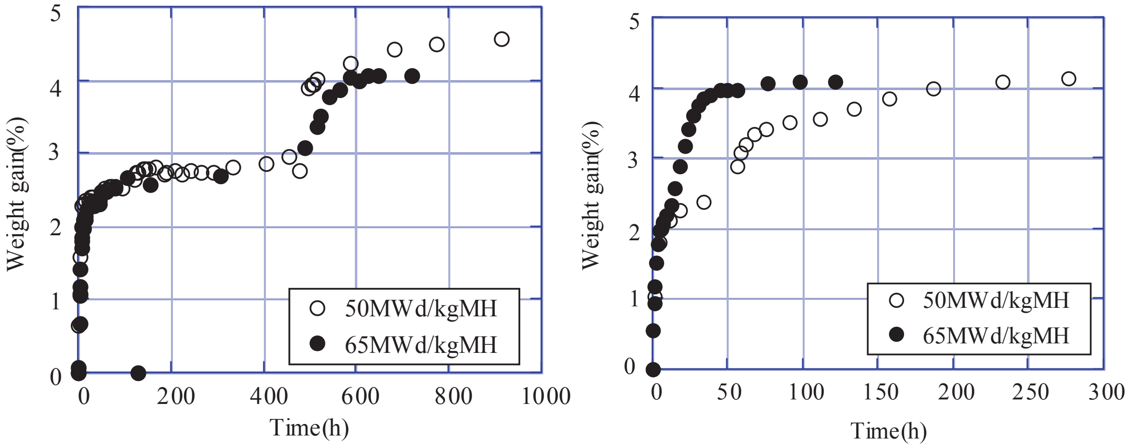
- Fuel Oxidation

In case of cladding with through-wall defects, air will come in contact with the fuel inside the rod. Oxidation of spent fuel in air is a two-step process of the form:

$UO_2 \rightarrow U_4O_9 \rightarrow U_3O_8$

In spent fuel oxidation experiments, the O/M ratios first approach a plateau near 2.4 [43]. The oxidation process proceeds according to the $UO_2 \rightarrow UO_{2.4}$ reaction, which does not result in appreciable fuel pellet density changes. Once the spent fuel oxidizes to $UO_{2.4}$, a plateau is reached where the fuel appears to 'resist' oxidation to higher oxides. Following this plateau, oxidation resumes after some time and continues until the U_3O_8 phase is reached. The transition from $UO_{2.4} \rightarrow U_3O_8$ results in a volume expansion of greater than 36% [44]. The increase in volume as spent fuel oxidizes to U_3O_8 results in high stress levels on cladding. Cladding rupture, or 'unzipping' may result. The $UO_{2.4} \rightarrow U_3O_8$ reaction is strongly dependent on both fuel temperature and fuel burnup.

UO_2 oxidation rates remain very low below ~250°C. If the temperature exceeds 250°C, UO_2 conversion to U_3O_8 becomes possible. To investigate the effect of burnup and temperature on oxidation behavior of a pellet, oxidation experiments were carried out on irradiated UO_2 fuels with burnups of 50 GWd/t U and 65 GWd/t U [45]. In these experiments, fuel samples were exposed to dry air at temperatures between 300 and 400°C. Figure 23 shows the weight gain curves at 300°C and 350°C. A weight gain of 4% corresponds to 100% formation from UO_2 to U_3O_8. At 300°C, there is no significant burnup difference in the oxidation behavior between 50 and 65 GWd/t U. On the other hand, at 350°C, the oxidation rate of the sample with a burnup of 65 GWd/t U is higher than that with a burnup of 50 GWd/t U beyond the O/M of 2.4. It has been speculated that the microstructure of the pellet affected oxidation behavior at 350°C.



(a) 300ºC (b) 350ºC

Fig. 23. Oxidation behaviour dependence on burnup and temperature.

(2) Thermal Creep

Spent fuel rods in dry storage undergo creep deformations at a rate determined by the internal fuel rod pressures, cladding temperatures, and composition and metallurgical conditions of the rod cladding. When assessing cladding integrity during dry storage, thermal creep has generally been evaluated as a potentially limiting degradation mechanism. Evaluation of the creep capacity of spent fuel rod claddings under long-term dry storage conditions requires the following information: (1) experimental creep data; including creep rupture criterion; and (2) an analytical model capable of simulating the cladding response under prescribed temperature and internal pressure conditions representative of dry storage applications.

Out-of-pile creep data for irradiated material were first generated by German researchers in the 1980s. Since then, several major programs, particularly in France and Japan, have resulted in a much larger set of data on irradiated materials. These data have led to the benchmarking of analytical models in the stress-temperature range of interest to dry storage applications.

Early implementation of a creep-limited approach in Germany led to a methodology that relies on the following considerations:

(1) Collection of an experimental database of creep test results of unirradiated cladding obtained under internal pressurisation conditions, which leads to a numerical model to predict creep strain .

(2) Conservative application of correlations obtained on unirradiated claddings to predict post-pile thermal creep of irradiated claddings, as it was shown by comparison of creep data of unirradiated and irradiated Zircaloys that creep of unirradiated material always describes post-pile creep conservatively (Fig. 24).

(3) Availability of a database of the total *strain capability* of fuel rod cladding under simulated dry storage conditions. For a burnup less than 40 GWd/t U, data from post-pile *burst test* indicate that cladding will reach 1% uniform strain before rupture. Because the rupture strain capability of the cladding is expected to decrease with burnup, this very conservative approach of using burst testing with its very high strain rates was replaced by accelerated testing with lower strain rates. Such a test program was conducted with spent fuel with burnup up to 64 GWd/t U [46] . The program provided the basis for the present German licence to store spent UO_2 and mixed-oxide fuel assemblies with average batch burnup up to 55 GWd/t U.

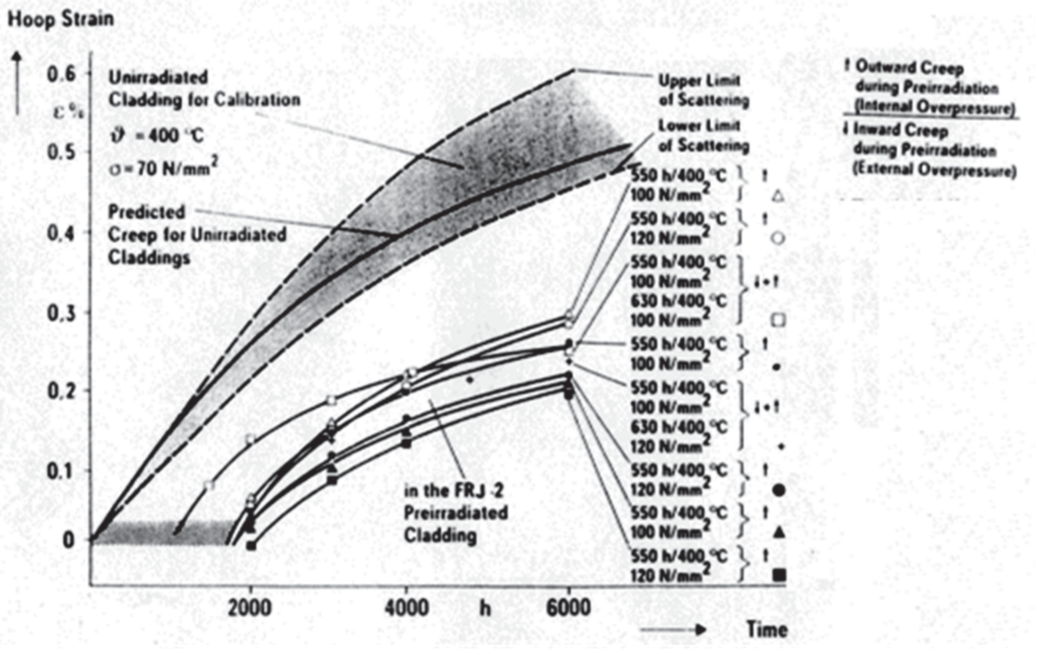


Fig. 24. Comparison of creep strain of irradiated and unirradiated Zry-4-cladding.

Although dry storage of commercial spent nuclear fuel is not an option presently envisioned in the French program, much work has been performed, both experimentally and analytically, in the context of the PRECCI (Programme de Recherches sur l'Evolution à long-terme des Colis de Combustibles Irradiés) project in order to better understand the long-term evolution of spent fuel under environmental conditions relevant to dry storage and geologic disposal. In particular, experimental data, derivation of creep rupture criteria and constitutive equations for cladding creep have been established. The work was pursued in the framework of the French laws for the management of nuclear wastes [47].

In Japan, creep behavior of fuel cladding is regarded as one of the most important issues for fuel integrity during long-term dry storage of spent fuel. A summary of the work performed by the Japan Nuclear Energy Organization (JNES) is presented in this section.

Creep deformation properties of unirradiated and irradiated BWR (Zry-2 irradiated up to 50 GWd/t) and PWR (Zry-4 irradiated up to 48 GWd/t) claddings have been investigated under various temperature and hoop stress conditions in two different controlling creep deformation regimes, respectively identified as grain boundary sliding and dislocation climb.

The creep strain is expressed as the sum of conventional primary creep and secondary creep:

$$\varepsilon = \varepsilon_p^s + \dot{\varepsilon}\, t \qquad \text{[Eq. 1]}$$

where:	ε	creep strain
	ε_p^s	saturated primary creep strain
	$\dot{\varepsilon}$	secondary creep rate
	t	time

The dependence of the secondary creep rate on stress and temperature is expressed by Eq. 2. The secondary creep rate is formulated as the sum of two creep mechanism contributions, as shown in Fig. 25.

$$\left.\begin{aligned} &\dot{\varepsilon} = \dot{\varepsilon}_L + \dot{\varepsilon}_H \\ &\dot{\varepsilon}_L = A_{SL}\left(\frac{E}{T}\right)\left(\frac{\sigma}{E}\right)^{n_{SL}} \exp\left(-\frac{Q_{SL}}{RT}\right) \\ &\dot{\varepsilon}_H = A_{SH}\left(\frac{E}{T}\right)\left(\frac{\sigma}{E}\right)^{n_{SH}} \exp\left(-\frac{Q_{SH}}{RT}\right) \end{aligned}\right\} \qquad \text{[Eq. 2]}$$

where:	$\dot{\varepsilon}$	secondary creep rate (1/hr)
	$\dot{\varepsilon}_L$	secondary creep rate in the low stress region
	$\dot{\varepsilon}_H$	secondary creep rate in the high stress region
	σ	hoop stress (MPa)

E	Young’s modulus (MPa)
R	*Gas constant (8.3144 J/mol*K)*
T	Temperature (K)
n_{SL}, n_{SH}	stress index of secondary creep rate
Q_{SL}, Q_{SH}	activation energy of secondary creep rate (J/mol)
A_{SL}, A_{SH}	Constant (K/MPa$*$hr)

Based on the same form of equation describing the secondary creep rate, the dependence of the saturated primary creep strain on hoop stress and temperature is expressed by Eq. 3.

$$\varepsilon_p^s = A_p\left(\frac{E}{T}\right)\left(\frac{\sigma}{E}\right)^{n_p}\exp\left(-\frac{Q_p}{RT}\right) \qquad \text{[Eq. 3]}$$

where:	ε_p^s	saturated primary creep strain
	n_P	stress index of saturated primary creep strain
	Q_p	activation energy of saturated primary creep strain (J/mol)
	A_p	constant (K/MPa)

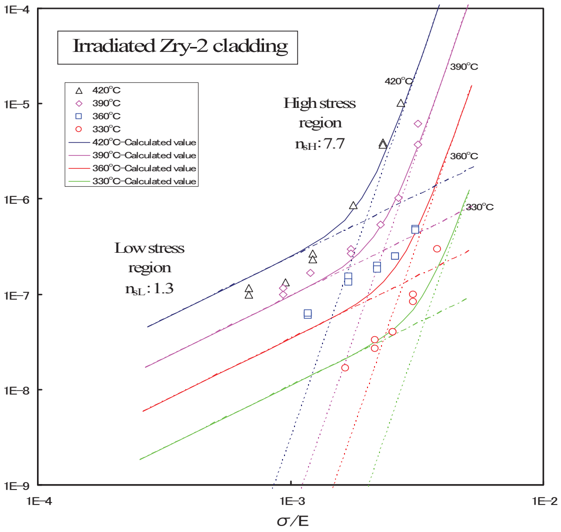


Fig. 25. Stress dependency of secondary creep rate.

In addition, since the creep strain of re-crystallized annealed (RXA) Zry-2 cladding is much smaller than that of stress-relieved material (SRA) Zry-4 cladding, the primary creep strain for Zry-2 cladding is represented by Eq. 4.

$$\varepsilon_p = \varepsilon_p^s(1-\exp(-rt^n)) \qquad \text{[Eq. 4]}$$

where:	ε_p	primary creep strain

ε_p^s	saturated primary creep strain
r	constant
t	time (h)
n	constant

From regression analyses of the experimental results, the following parameters are obtained:

For irradiated Zry-4 cladding,

$A_{SL} = 4.04\times10^{1}$ K/MPa/h: $n_{SL} = 0.48$; $Q_{SL} = 109.9\times10^{3}$ J/mol

$A_{SH} = 2.50\times10^{35}$ K/MPa/h; $n_{SH} = 7.39$; $Q_{SH} = 297.7\times10^{3}$ J/mol

$A_P = 6.58\times10^{4}$ K/MPa; $n_P = 1.29$; $Q_P = 77.2\times10^{3}$ J/mol

For irradiated Zry-2 cladding,

$A_{SL}=8.12\times10^{-2}$ K/Pa/h; $n_{SL}=1.3$; $Q_{SL}=1.28\times10^{5}$ J/mol

$A_{SH}=2.41\times10^{26}$ K/Pa/h; $n_{SH}=7.7$; $Q_{SH}=2.63\times10^{5}$ J/mol

$A_p=8.33\times10^{-2}$ K/Pa; $n_p=1.3$; $Q_p=7.71\times10^{4}$ J/mol

For $\varepsilon_p^s < 5.70\times10^{-3}$, $r=1.64\times10^{-1}$ and $n=4.65\times10^{-1}$

The secondary creep rate and creep strain of irradiated cladding were, for all temperature conditions and stress regions, smaller than those of unirradiated cladding, which confirms the creep reduction effect due to irradiation damage within the range of test conditions. The effect was smaller at lower temperature and lower stress. Assuming the creep mechanisms to be dislocation climb and grain boundary sliding in the high and low stress region, respectively, stable irradiation defects such as point defect clusters, dislocation loops, etc. prevent dislocation movement; as a result, creep suppression becomes more apparent in the high stress regions where creep by dislocation climb is predominant. On the other hand, because the irradiation damage is not considered to be very effective in suppressing creep due to grain boundary sliding, the suppressive effects in the low stress regions are relatively small.

The hydrogen content in the cladding increases during in-reactor service. For the tests conducted on *unirradiated* cladding, the effects of hydrogen on cladding creep behavior in the investigated range of temperature and hoop stress were as follows:

When the hydrogen content is below the solubility limit, for BWR Zry-2 cladding, creep showed a slight acceleration by charged hydrogen, while no significant tendencies were observed for PWR Zry-4 cladding. When the hydrogen content is above the solubility limit, creep suppressive effects increased with increased hydrogen content in PWR cladding, while, for BWR cladding, a slight accelerating effect was observed in low stress region, and suppressive effects were observed in the high stress region. No effects of hydrides reoriented in the radial direction were observed on creep rate.

The threshold creep strain marking the transition from secondary to tertiary creep was investigated. The threshold strain for transition to tertiary creep is likely to be more than 10 % for unirradiated cladding and 1% for irradiated cladding in the high stress region. Threshold creep strain for transition into tertiary creep was not investigated in the low stress region as it would have required obtaining creep data over much longer times.

In support of interactions between the US utility industry and the U.S. Nuclear Regulatory Commission, EPRI developed a creep-based methodology for high-burnup fuel placed in dry storage [48]. Behavioral regimes considered included the effects of irradiation-damage recovery and hydrogen content on the cladding creep rate, the effects of outer-surface corrosion including oxide spallation on cladding stress localization and its consequent effect on creep deformations, and the role of temperature and stress decay on the potential for creep rupture. Application to available creep-rupture tests showed that creep rupture is a stress-induced plastic instability as a terminal state of tertiary creep, which is highly unlikely to occur in dry storage of spent light-water reactor fuel consisting of low-enriched uranium. This is because cladding creep deformations under dry storage conditions are self-limiting, characterized by a continuously decreasing strain rate due to a continuously decreasing cladding stress in proportion to the creep-induced volume expansion and decreasing temperature. As a result, it was shown that, during dry storage, the creep strain is unlikely to exceed the critical strain domain leading to plastic instability.

Concurrently, experiments carried by Argonne National Laboratories demonstrated the creep capacity of Zircaloy-4 claddings from Surry (~35 GWd/MTU) and H.B. Robinson (~68 GWd/MTU). Diametral creep strains as high as 8.5% (Surry) and 5.5% (H.B. Robinson) were measured with no indication of local reductions in cladding thickness.

An example of analytical simulation of dry storage for a high-burnup fuel rod with oxide thickness of 120 μm, and a hydride lens of depth equal to 50% of original thickness is shown in Fig. 26. The rod initial temperature of 400°C and internal pressure of 19.1 MPa are shown on the figures. The temperature of 440°C is the peak temperature during a 24-hour drying cycle, including heat-up and cool-down period of 8 hours each. Figure 26 shows the hoop strain in the hydride lens ligament (thin section) and away from the lens (thick section). The oxide thickness is treated as an equivalent metal loss. Figure 26 also shows the self-limiting behavior of cladding creep (even in the local hydride lens region).

An important behavior characteristic that is not simulated in pressurized-tube creep tests is the effect of fuel pellets on constraining the creep deformations of high-burnup fuel rods containing localized hydrides. Because of the presence of a hydride lens in a localized region of the cladding, the creep deformations are not uniform and, further, the deformations tend to localize in the hydride-lens ligament. This causes the cladding to be pulled inward at a position 90 degrees away from the lens region, further increasing the deformations in the thinned ligament. The fuel pellets prevent this behavior from developing and constrain the deformations in the hydride-lens region. This mode of behavior is absent in an empty tube test, as illustrated in Fig. 27. The difference between the two behavior modes is quite significant.

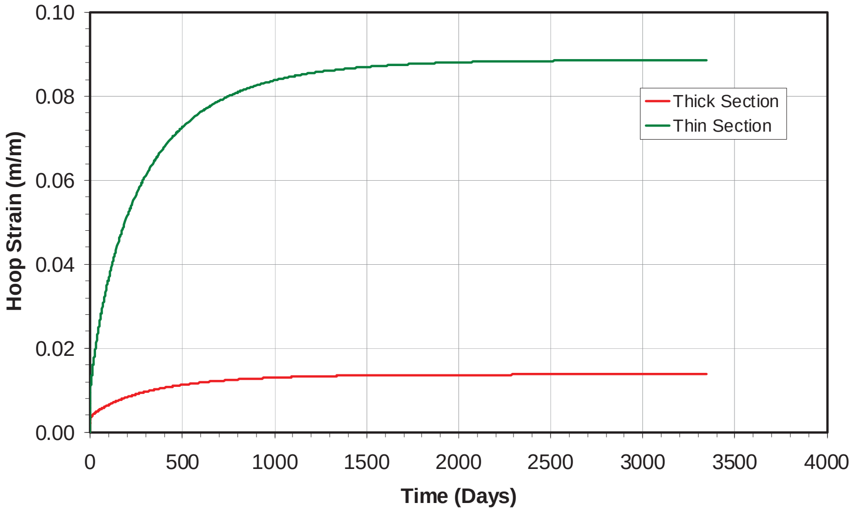


Fig. 26. Strain history for a fuel rod in a dry cask subjected to a 24-hour drying cycle with a peak temperature of 440°C.

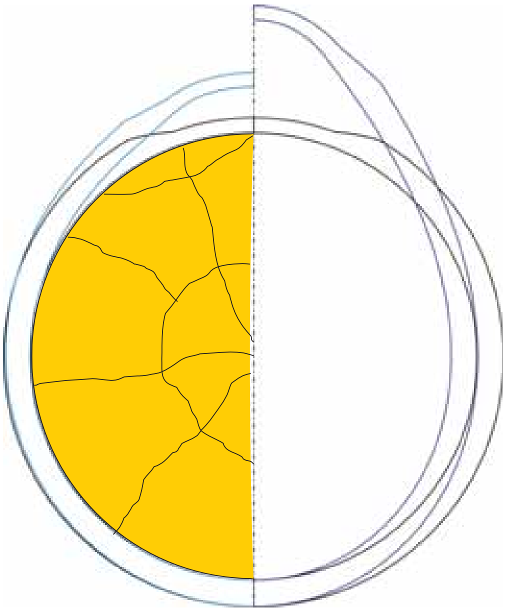


Fig. 27. Comparison of the creep response of a fuel rod (left side) with the empty tube creep test simulation of Fig. 26 (right side), illustrating the constraining effect of fuel pellets on the local creep deformations.

The result of these investigations resulted in a determination by the US Nuclear Regulatory Commission summarized as follows:

> *'In general, these data and analyses support the conclusions that: (1) deformation caused by creep will proceed slowly over time and will decrease the rod pressure; (2) the decreasing cladding temperature also decreases the hoop stress, and this too will slow the creep rate so that during later stages of dry storage, further creep deformation will become exceedingly small; and (3) in the unlikely event that a breach of the cladding due to creep occurs, it is believed that this will not result in gross rupture. Based on these conclusions, the NRC staff has reasonable assurance that creep under normal conditions of storage 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 (752°F). As discussed below, this temperature will also limit the amount of radially oriented hydrides that may form under normal conditions of storage'* [50].

The Spanish organizations CSN, ENRESA and ENUSA have successfully carried out a program to generate thermal creep data on high burnup ZIRLO (68 GWd/MTU) irradiated in the Spanish Vandellos nuclear power plant [51]. The creep results were consistent with the expected behaviour from reference CWSR Zircaloy-4 material. During one test, a specimen leaked at an engineering strain of the tube section of 17%; remarkably, the crack closed due to de-pressurization, indicating that the material retained significant ductility.

(3) Stress Corrosion Cracking

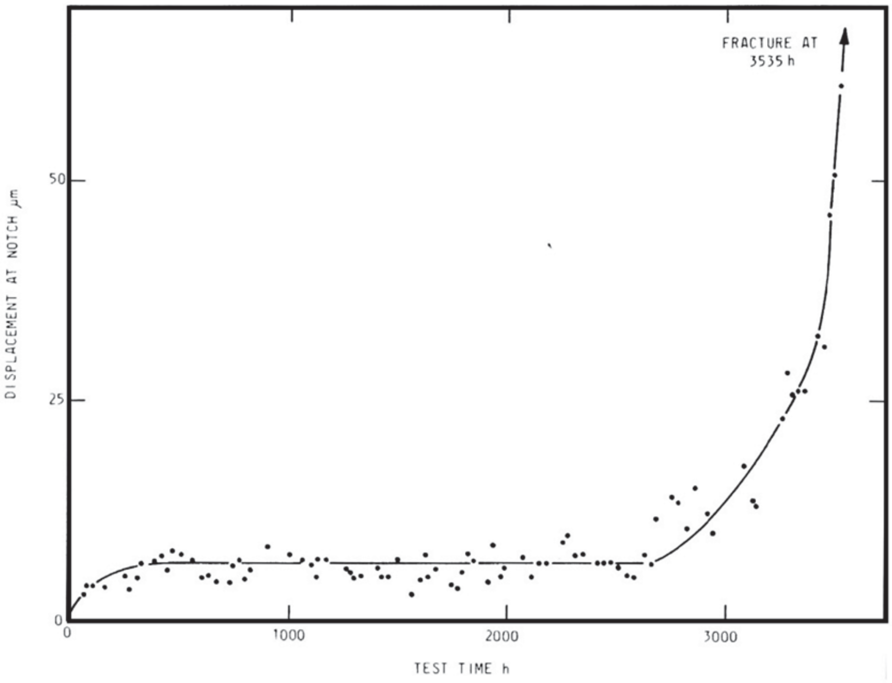
Iodine-induced stress corrosion cracking (SCC) occurs only within a specific high temperature range, in the presence of chemically active iodine and adequate stresses. Fission product behaviour can be characterized by a high stability against release. UO_2-fuel crystallizes in a CaF_2-type lattice and contains a plurality of vacancies, which aid in the retention of fission products generated by reactor operation. Also, each fissioned U-atom leaves an additional vacancy in the lattice. As a result, even in high burnup fuels, not all vacancies are occupied, and the UO_2-crystal has sufficient capability to retain the fission products. A complete discussion of this topic is available in Peehs et al. [41]. In summary, the fission products generated in the UO_2-fuel under in-service conditions are practically immobile in the UO_2-fuel lattice in the temperature ranges typical of interim storage, and iodine is not present in a form that could trigger SCC.

As the combination of SCC agent and stress conditions required for crack propagation are normally absent, it can, therefore, be concluded that cladding failure via this mechanism is not expected to occur. Consequently all fission-product-driven defect mechanisms such as stress corrosion cracking, uniform fuel rod internal fission product corrosion of the cladding, and localized fuel rod internal fission product corrosion of the cladding are not active.

(4) Delayed hydride Cracking (DHC)

There are many ways in which hydrogen-induced cracking of zirconium-based alloys can occur. DHC is a very specific mechanistic process requiring tri-axial stressing of the zirconium to dilate the crystal lattice. Tri-axial stresses dilate the Zr lattice tetrahedral sites that are the ones occupied by hydrogen atoms in solution. This allows the hydrogen atoms to diffuse up the stress gradient and precipitate in the peak stress region. Hydrides grow in this region by dissolving hydrogen in adjacent hydrides in unstressed regions. The hydrides grow until big enough to crack. Although DHC has been observed in Zircaloy specimens that are sufficiently thick, DHC is not expected to be an active degradation mechanism in cladding tubes, given that the latter do not appear to have enough wall thickness to generate much tri-axial stress.

Laboratory tests of notched and pre-cracked specimens show that the process evolves in two stages. Stage-I is comprised of crack initiation followed by a rapid increase in crack velocity, of one or two orders of magnitude, until it transitions to the stable crack growth Stage-II. The failure time is determined primarily by the nearly constant crack growth velocity of Stage-II. During Stage-II, under isothermal conditions, the hydrogen in solution diffuses to the high stress zone surrounding the crack tip. With the continuous hydrogen diffusion to the crack tip region, the local hydrogen concentration rises until it exceeds the solubility limit and hydrides begin to precipitate ahead of the crack tip in the form of platelets oriented perpendicular to the direction of the stress. When the hydride reaches a critical size, it fractures, allowing the crack to advance until it is arrested in the tougher Zircaloy material past the hydride, where a new highly stressed crack tip is formed, and the process is repeated. The crack continues to propagate in this intermittent manner until failure by plastic instability of the remaining ligament. This process is illustrated in Fig. 28.



Displacement-time curve for round notched bar of Zircaloy-2 containing 100 ppm hydrogen and tested at 350 K with K_I of 15 MPa $\sqrt{m}$.

Fig. 28. Intermittent DHC crack evolution.

Rashid et al. [52] present an analysis defining five conditions that should exist for DHC to be an active mechanism in LWR fuel during dry storage. The analysis concludes that DHC is not an operative mechanism in dry storage. It shows that stress intensities in the LWR fuel cladding are below the critical stress intensities for either crack initiation or sustaining crack growth.

However, DHC is considered to be the most critical mechanism that could affect the CANDU bundle integrity during dry storage (see later section).

(5) Hydride re-orientation

An important issue in the safety evaluation of spent fuel during storage and transportation is the effect of hydrides on cladding resistance to failure. At issue is the safety consequence of loading challenges such as accidental impact during handling, cask tip-over during earthquakes, or accidental drop during transportation. The degree to which spent fuel cladding, after a number of years in dry storage, can retain sufficient strength and ductility to resist catastrophic failures under these loading challenges would depend, among several factors, on the spatial distribution and orientation of the hydrides. The cladding tube manufacturing process generates a texture in the cladding that causes the hydride-platelets to precipitate in a predominantly tangential orientation. At the end of the active fuel cycle in PWRs, the total hydrogen concentration in Zircaloy-4 cladding can typically be as high as

500-600 ppm, which is predominantly in the form of circumferentially oriented zirconium hydride platelets, with radially varying spacing. This hydride morphology evolves very slowly during reactor operation as a consequence of hydrogen uptake during the slow waterside corrosion process, causing significant, but operationally tolerable, changes in the cladding mechanical properties. However, when spent fuel is placed in dry storage, operationally induced cycles of hydride dissolution/re-precipitation may be activated, which can lead to a mixed, circumferential and radial, hydride structure, depending on temperature and cladding hoop stress histories. Such a hydride structure can have significant effect on cladding mechanical properties and failure limits, especially in the lower range of temperatures (<150°C).

Fuel specifications for German reactors limit cladding hoop stresses to or less than 120 MPa.

Spent-fuel-cladding temperature reaches its highest level during cask drying, and is typically bounded by the regulatory limit of 400°C, which was specifically chosen to limit re-orientation of hydrides to an acceptable level. This temperature limit effectively sets the initial conditions for the fuel rod internal pressure, which defines the initial hoop stress in the cladding, and the amount of hydrogen in solid solution that can potentially re-precipitate in the form of zirconium hydrides as the cladding slowly cools down under stress during dry storage. At the initial temperature of 400°C the total amount of hydrogen in solid solution in Zircaloy-4 cladding is ~200 ppm, which is the maximum amount of hydrogen that can potentially precipitate, partly as circumferential hydrides and partly as radial hydrides, with the latter having the greater effect on cladding behavior. The precipitation rate and the concentration fractions of precipitating radial and circumferential hydrides depend on the initial magnitudes and the time histories of temperature and stress during storage. Precise determination of the magnitude of radial hydrides as function of storage time is an essential pre-requisite for the development of cladding failure criteria. To this end, a hydride precipitation model was developed [53].

Knowing the initial hydrogen content of the cladding and the time histories of the temperature and stress, the model calculates the circumferential and radial hydride concentrations as they evolve with time. The model was verified and validated using hydride precipitation data obtained for specimens cooled from various initial temperatures under constant stress to room temperature. A unique attribute of the model, for which no data currently exist, is that it can describe the hydride structure at any intermediate temperature considering time-varying stress typical of spent fuel dry storage.

The starting points for the model are the mathematical expressions developed by Ells [54], which yields the radial hydride fraction when observation is made at room temperature after cooling from an initial temperature under constant stress. However, these expressions cannot be applied to continuous cooling where both the temperature and stress are varying with time, as is the case during dry storage. Therefore, an expression for the precipitation rate as a continuous function of time-varying stress and temperature, which can then be integrated to provide the radial hydride concentration at any observation time, was derived.

As an example, a case with an initial stress of 150 MPa, which is a conservative estimate of cladding stress of high-burnup spent fuel at 400°C, and a maximum temperature of 400°C is considered. The temperature history in the cladding under decay power in high-burnup fuel is approximated by the following equation,

$$T = 517.48 + 155.52 \exp(-0.0877\ t) - 0.6922575\ t$$

where t is expressed in years and T is in Kelvin.

The following expression for the cladding hoop stress is chosen,

$$\sigma(t) = \sigma_0 \; T(t) \,/\, T^{initial}$$

where σ_0 stands for the initial hoop stress.

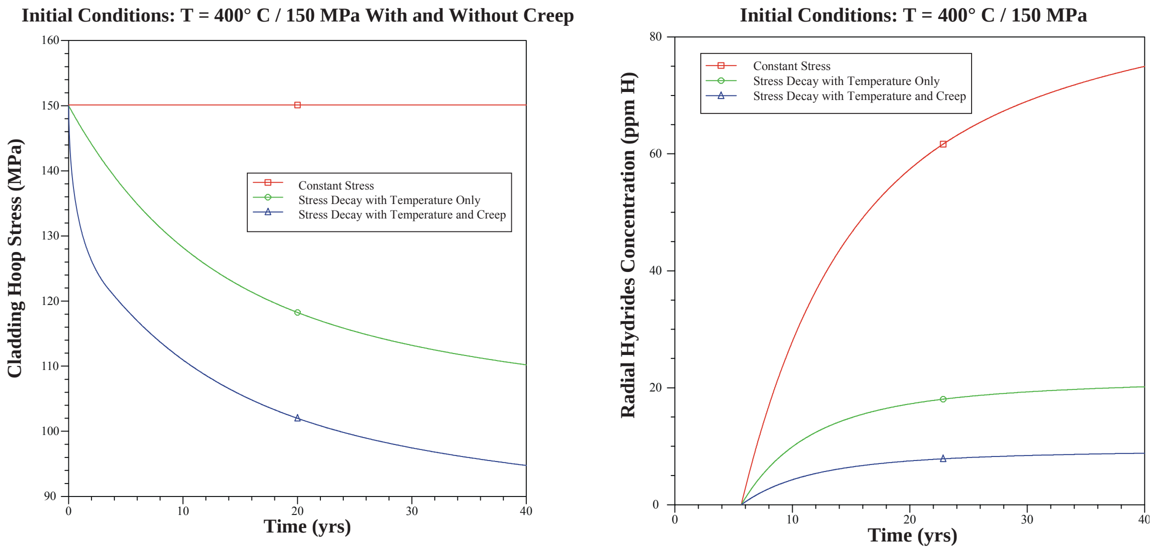
Figure 29 shows the results for 40 years of dry storage, considering three stress histories: constant stress, decaying stress in proportion to temperature decay only, i.e., ignoring the effects of cladding creep on fuel rod void volume, and considering the creep effects as would normally occur during dry storage. The three stress histories are shown in Fig. 29 (left-side graph), and Fig. 29 (right-side graph) shows the corresponding radial hydrides concentrations at the end of 40 years of dry storage where the cladding temperature has dropped to ~220°C. Hydride precipitation does not begin until approximately year-6 of storage, which is the period of time needed for the temperature to drop to ~335°C, which is the temperature corresponding to the onset of re-precipitation according to the hydrogen solubility upon cooling [55].

The most realistic result in is depicted by the lowest curve in Fig. 29 (right-side graph). In a fuel rod, which is a closed system with constant gas mass, cladding creep takes place under continuously decreasing stress because of decreasing temperature and expanding void volume. After a relatively short period of time in dry storage, usually about five years, depending upon the initial stress and temperature, cladding creep virtually stops due to the combined effect of reduced temperature and stress on the creep rate. The model's application to dry storage shows that only a small fraction of the hydrides in high burnup cladding can become aligned radially. As depicted in Fig. 29 for the constant-stress case, the amount of hydrogen that could potentially precipitate after 40 years in the form of radial hydrides is predicted by the model to be a small fraction, less than 15 ppm, of a typical hydrogen content of ~ 600 ppm in high burnup cladding. The constant-stress case is considered physically unrealistic since there is no known mechanism by which the cladding hoop stress can remain constant during dry storage. Even if one ignores the effects of creep the stress is expected to follow the temperature decay, in which case the model predicts that the maximum concentration of radial hydrides is about 20 ppm.

In Gruss et al. [50], the US Nuclear Regulatory position was summarized as follows:

> *'In general, a temperature limit of 400°C that is specified for normal conditions of storage and for short-term fuel loading and storage operations (which includes drying, backfilling with inert gas, and transfer of the cask to the storage pad) will limit cladding hoop stresses and limit the amount of soluble hydrogen available to form radial hydrides. The use of a 400°C temperature limit for normal conditions of storage and for short-term fuel loading and storage operations will simplify the calculations in Safety Analysis Reports (SARs) while assuring that hydride reorientation will be minimized.*
>
> *It should be noted that during normal conditions of storage there will be a distribution of cladding temperatures less than the maximum allowable cladding temperature resulting in distributions in the rod internal pressures and the cladding hoop stresses in any one dry cask storage system (DCCS). In general, the maximum allowable temperature will be 400_C or the maximum allowable*

temperature specified by the applicant. It is expected that due to these distributions only a small fraction of the rods will experience the temperature and stress conditions that could lead to the formation of radial hydrides during normal conditions of storage.'



Stress History During Dry Storage

Radial Hydrides Concentration During Dry Storage

Fig. 29 Evolution of radial hydrides (right figure) under three stress histories (left figure) [The lower curves represent the most realistic evolutions for actual dry storage conditions].

Since 2004, the Japan Nuclear Energy Safety Organization (JNES) has been performing tests, using irradiated claddings (from spent BWR and PWR fuel with burnup up to 55 GWd/MTU), in which the effects of hydride reorientation are investigated.

The main cladding tube materials examined in the study were Zry-2 cladding from BWR spent fuel, Zry-4 cladding from PWR spent fuel, and two types of advanced alloy, MDA and ZIRLO cladding from PWR spent fuel. Unirradiated BWR Zry-2 hydrogen-charged specimens were also tested.

Figure 30 shows the metallography of BWR Zry-2 cladding before and after hydride reorientation testing (HRT).

- Compared to (a), specimens (b) and (g) show a small degree of hydride reorientation into the radial direction after HRT with a hoop stress of 70 MPa starting from a temperature of 300°C, and a cooling rate of 30ºC/h; hydride reorientation was observed more clearly in the inner side area of the cladding wall near the Zr liner, and little reorientation seemed to occur in the area near the outer surface. Significant amount of hydride reorientation was observed for specimens after HRT at 400°C; at that temperature, all the hydrogen was considered to be dissolved completely for most of the specimens.

- The hydrogen content of unirradiated cladding specimens were so low that the hydrogen dissolved completely at each HRT temperature. Comparing specimens (c) and (f), the reorientation for irradiated cladding seemed somewhat larger than that for unirradiated cladding.

- Specimens g to j show the effect of cooling rate. The amount of hydride near the inner surface area of Zry-2 matrix was decreased and the amount of hydride in the Zr liner was increased with decreasing cooling rate. This phenomenon about cooling rate effect on Zr-lined cladding was the result of hydrogen migration from the Zry-2 matrix to the Zr liner due to the terminal solid solubility precipitation (TSSp) difference between Zr and Zry-2 [56]. A lower cooling rate allows more time for hydrogen diffusion to the Zr liner from Zry-2. On the other hand, Zry-2 cladding with no Zr liner showed a relatively large degree of hydride reorientation, as observed in specimen j.

The results of ring compression tests for BWR Zry-2 cladding with a Zr liner are summarized in Fig. 31. The crosshead displacement ratio, which can be considered as a ductility index, did not show the particular hoop stress dependence for the HRT 300°C and 250°C specimens, although some degree of reorientation was observed for HRT 300ºC, 70 – 100 MPa specimens. On the other hand, the crosshead displacement ratio increased for HRT 400°C, 0 MPa (only heat treatment with no stress) specimens compared to as-irradiated specimens, and it decreased with the increase of hoop stress in HRT, or the amount of radial hydride.

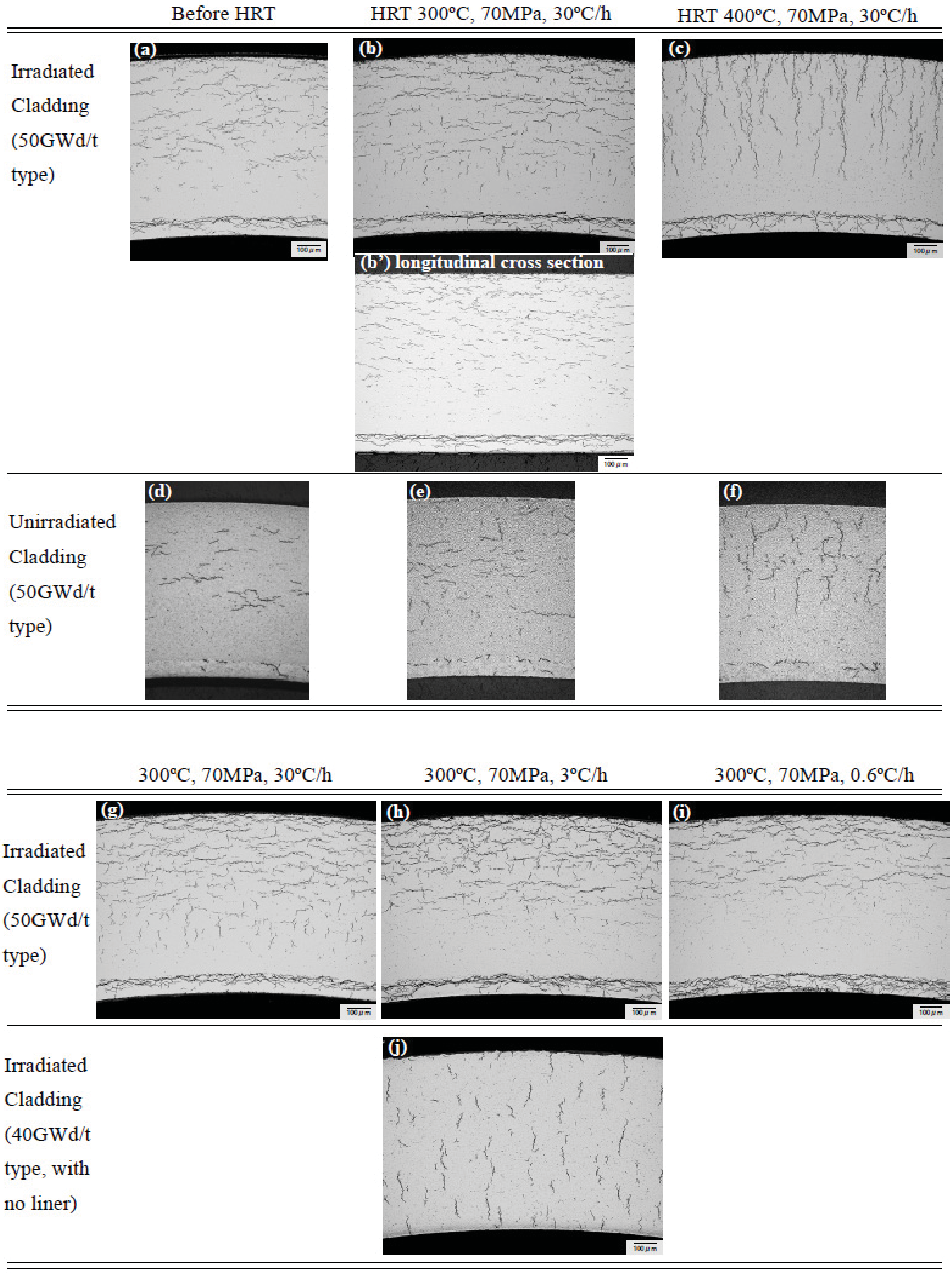


Fig. 30. Metallography of BWR Zry-2 cladding specimens in hydride reorientation test [radial cross section except for (b')].

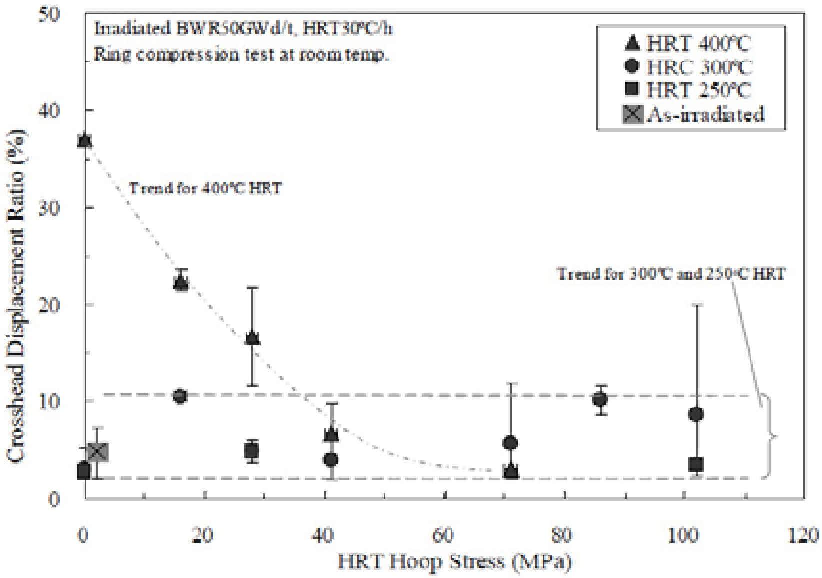


Fig. 31. Correlation between the Ductility of Specimens and HRT Conditions for Irradiated BWR Zry-2 Cladding with Zr Liner.

For irradiated BWR Zry-2 cladding, significant hydride reorientation to the radial direction occurred under the relatively low hoop stress and the temperature conditions in HRT, such as less than 70 MPa, 400ºC. The degree of reorientation depended on the HRT solution temperature rather than the estimated temperature at which the hydride precipitation occurred. It is suggested that reorientation behavior is a complex function resulting from several features, including the effects of precipitated (not dissolved at HRT temp.) hydride and the memory effect in relatively moderate conditions such as 300ºC, 70 MPa, 30ºC/h HRT. In relatively low cooling rate HRT, the hydride preferential precipitation in Zr liner increase for Zr lined cladding compared to that in Zr liner with a relatively high cooling rate. The ductility of the cladding specimens after 400°C, 0 MPa, 30ºC/h HRT increased in ring compression testing at room temperature compared to no HRT (as-irradiated) specimens, as recovery of irradiation damage at 400ºC due to annealing occurred, which affected the ductility of the irradiated Zry-2 cladding. The cladding with no Zr liner showed a high degree of reorientation in 300ºC, 70 MPa, 3ºC/h HRT, and the lowest ductility among the BWR specimens in ring compression test after the HRT, although the hydrogen content of no-liner specimens was the lowest (40 ppm H) among the tested materials.

The results of hydride reorientation tests for irradiated PWR 48 GWd/t type Zry-4 cladding were evaluated in the same way as for BWR cladding; the results are shown in Fig. 32. The hydrides in the mid-wall area were used, because the hydride reorientation behavior was clearer and the hydride morphology in the mid-wall area had the higher correlation to the ductility, compared to that in an area that included the hydride rim. The ratio of radial hydride increased for the specimens after HRT 115 MPa, 300°C, 30ºC/h compared to as-irradiated specimens. The effect of cooling rate on hydride morphology at 30ºC/hr compared to 3ºC/hr was noticeable, but was smaller at 3ºC/hr compared to 0.6ºC/hr.

The results of ring compression test for 48 GWd/t type Zry-4 cladding after HRT are summarized in Fig. 33. The crosshead displacement ratio was almost the same level for the specimen after HRT 250ºC, 100 MPa and 275ºC, 100 MPa compared to as-irradiated specimens. The specimens after HRT 340ºC, 100 MPa and 300ºC, 100 MPa showed lower crosshead displacement ratios compared to as-irradiated specimens.

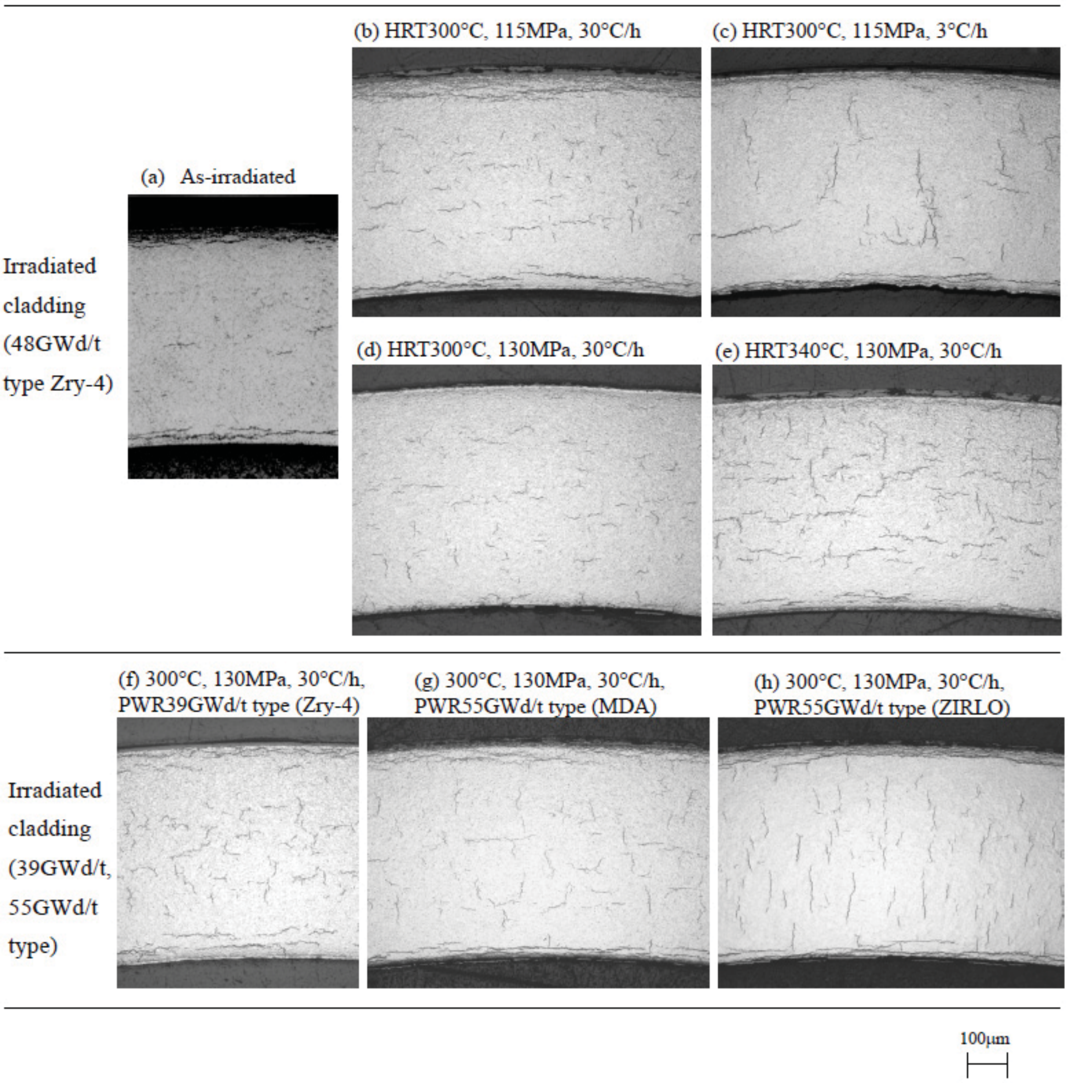


Fig. 32. Metallography of PWR cladding specimens before and after HRT.

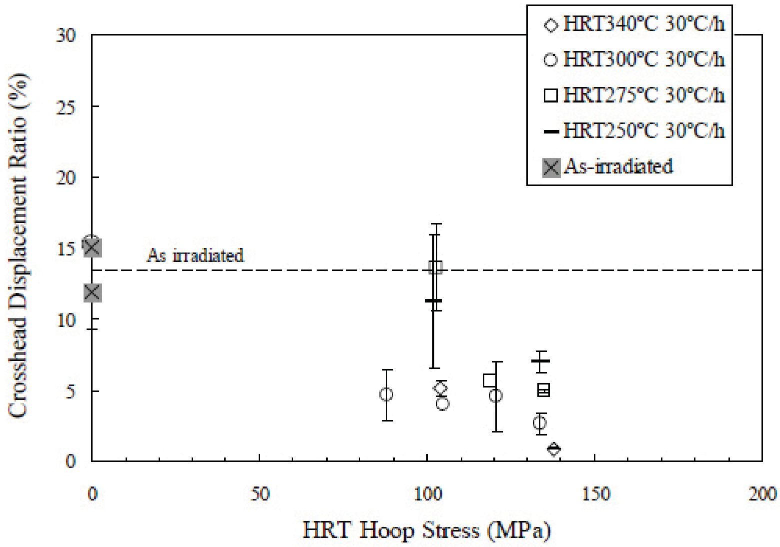


Fig. 33. Correlation between the ductility of specimens and the HRT conditions for irradiated PWR 48-GWd/t Zry-4 cladding.

For irradiated PWR Zry-4 and improved zirconium alloy cladding, little increase of the radial hydride ratio was observed in HRT with 100 MPa, 340ºC or less. On the other hand, the amount and the length of the hydride in the mid-wall area was a function of temperature and cooling rate in HRT due to the hydrogen migration from the hydride rim area. The ductility in ring compression deformation was affected by the orientation, amount, and length of hydride in the mid-wall area.

(6) Hydrogen Migration and Re-distribution

Given the pickup of hydrogen by the cladding during irradiation, hydrogen can migrate from the high temperature to the low temperature region of the cladding as a result of the axial temperature profile of the fuel rod [57]. Hydrogen may precipitate in the colder regions in the form of hydrides, and consequently, lead to increased brittleness, which may affect fuel integrity.

Hydrogen redistribution experiments in the axial direction of cladding tube were carried out and the data related to hydrogen migration such as heat of transport, hydrogen diffusion coefficient, and solubility limit were obtained [60]. Samples from twenty-year dry-stored spent PWR-UO_2 fuel rods, whose burn-ups were 58 GWd/t U (Sample-1AR) and 31 GWd/t U (Sample-1BR), were used in the hydrogen redistribution experiment. Unirradiated claddings were also used as reference materials. Samples of 30-mm length was mounted between heaters, and surrounded with thermal insulators. The temperature on one end of the sample was kept at 653K and that on the other end was kept at 533 K. The atmosphere during the experiment was the air in order to keep an oxide layer on the cladding surface to prevent hydrogen from escaping out of the sample (Fig. 34).

Figure 35 shows the hydrogen concentration analysed on each specimen after heating for ten days. Hydrogen on twenty-year stored cladding migrated from the high temperature to the low temperature region in the samples. Before the test, the difference in hydrogen concentration of adjacent specimens was less than 2.2 ppm and hydrogen profiles in the samples were regarded as almost flat. From these results, the heat of transport, diffusion coefficient and solubility limit of hydrogen were calculated by best fitting using a time-dependent hydrogen diffusion equation [57].

The hydrogen redistribution in the PWR-UO_2 rod after forty years of storage was then estimated by a one-dimensional hydrogen diffusion calculation using the heat of transport, the diffusion coefficient and the solubility limit obtained in the study [60]. Figure 36 shows the calculated hydrogen redistribution in a fuel rod after forty years of storage. The axial hydrogen migration is not significant after forty years of storage under helium atmosphere, due to the low value of the hydrogen diffusion coefficient, except at fuel rod ends. Hence the hydrogen profile was not significantly different from the initial one. It is, therefore, concluded that hydrogen migration has no significant effects on fuel cladding integrity during dry storage.

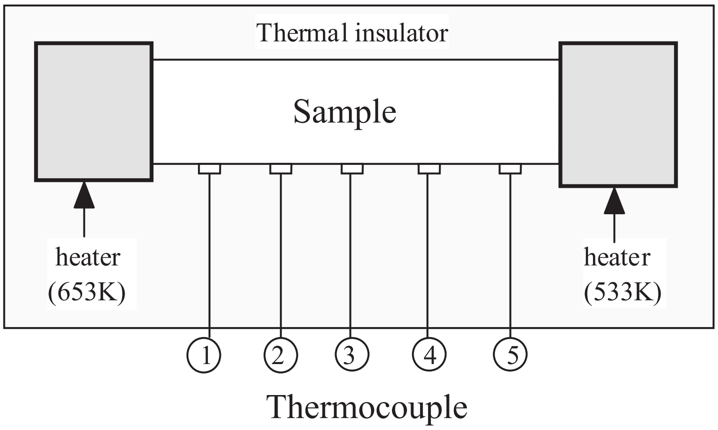


Fig. 34. Apparatus used for the hydrogen redistribution experiment.

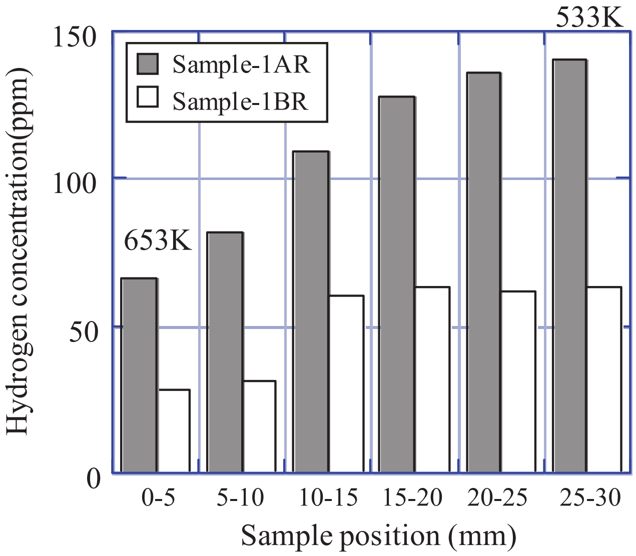


Fig. 35. Hydrogen distribution in samples after the 10-day redistribution experiment.

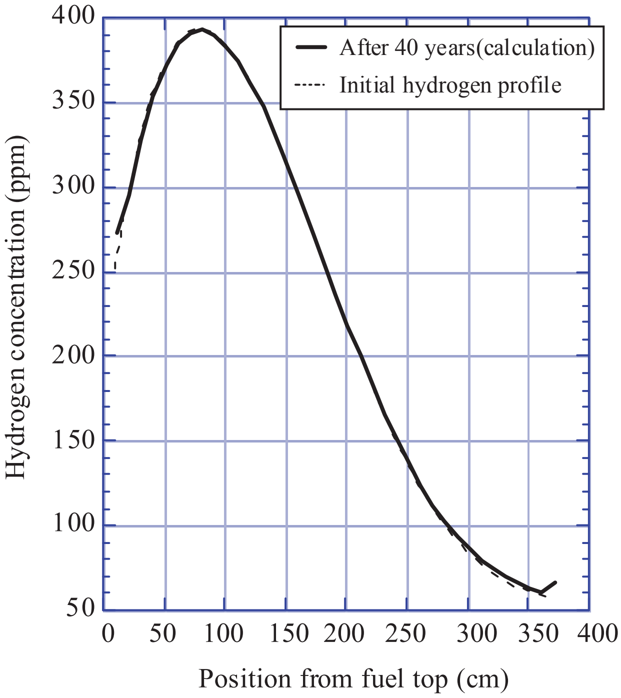


Fig. 36. Axial hydrogen redistribution after 40 year of storage (calculation).

7.2.2. AGR

The cladding material used in the manufacture of AGR fuel pins is 20Cr/25/Ni, which is a Nb-stabilised stainless steel. During reactor operation, stainless steel components in the 340-520°C temperature range (peak at 420°C) become sensitised, primarily as a result of radiation induced segregation. The cladding of elements (1-3) of the seven stack (usual) fuel stringer, are affected by this mechanism. Sensitisation, coupled with stress induced at the anti-stacking grooves of the fuel pin, can result in cladding failures for pins stored in moist air conditions or in pond storage environments (with high chloride ion concentrations); through the failure mechanism known as irradiation-induced intergranular stress corrosion cracking (IGA). This has implications for potential air ingress at reactor shut downs, initial cooling/storage at reactor in dry buffer storage tubes and interim storage ponds.

For short to medium term fuel storage durations, IGA is the prominent defect mechanism for AGR fuel cladding. Unlike Zr-based alloys, if the storage durations become prolonged (i.e., hundreds of years) general waterside corrosion becomes a significant factor, especially where initial in-reactor cladding wall thinning has been significant. This failure mechanism also affects the fuel element stainless steel structural materials (braces etc.); in this case, it is most dominant in the materials of fuel elements 3-5 of the stringer.

Sensitisation of stainless steel is a widely recognised and researched topic. It is caused by the free carbon in the alloy, reacting with the chromium-alloy, to form chromium carbides on the metal grain boundaries. The formation of chromium-rich carbides on the grain boundaries denudes them of the chromium alloying additions and the stainless steel considered becomes sensitised to localised grain boundary corrosion in environments, where the bulk material is passive.

However, the specification of AGR cladding steel is set to avoid chromium carbide precipitation by firstly including a low carbon limit and secondly by inclusion of niobium.

The niobium constituent is there specifically to stabilise the steel against the precipitation of chromium carbides, by allowing preferential reaction of carbon with niobium. A 30-minute 930°C anneal allows this preferential reaction to occur and the niobium concentration is specified to ensure an excess over (carbon + nitrogen). The concentration of carbon left in solution in the steel at the start of irradiation is thus very low. Clearly, the carbon deposit on the coolant side of AGR cladding is a potential source of carbon, but diffusion of carbon was not considered fast enough to be able to sensitise material at depth in the cladding in the relevant temperature range; the inability of carbon to diffuse deep into the cladding at the relevant temperatures has been confirmed experimentally [61].

Radiation-induced segregation, however, has been found to be the over-riding factors in AGR cladding sensitisation. Fast neutrons are capable of generating defects in metals by 'non-elastic' displacement of atoms. An atom can be knocked from its normal lattice site into an interstitial location and a vacancy created. Vacancies are mobile and can be removed from the lattice at sinks. A grain boundary is a good sink. In moving in the lattice, a vacancy needs to exchange positions with another atom. The diffusion coefficients of iron, chromium and nickel in an austenitic steel are such that chromium diffuses fastest. There is thus a slight bias in favour of vacancies exchanging with chromium as they move towards a sink. The net effect is to move chromium away from the grain boundary. Nickel diffuses the most slowly of the three main steel constituents, and, as a result, nickel tends to become enriched at grain boundaries.

Chromium-depleted zones on grain boundaries can only be maintained if the supply of chromium from within the grain is not fast enough to replenish the boundary region. When vacancy mobility is very low, very few vacancies will reach sinks. Thus a window in irradiation temperature exists, above and below which irradiation-induced sensitisation is not observed. Some important points follow. First with continued irradiation at constant neutron flux, there will be a move towards a steady-state chromium depletion profile at a grain boundary. The steady state results from a balance between vacancy migration rate (number and mobility) and chromium diffusion rate (gradient and mobility). Based on the modelling of experimental observations, predictions were made of the likely AGR fuel burn-ups at which radiation induced sensitisation might level off. Second, the nature of the chromium depletion profile at a grain boundary (width of depleted zone and reduced chromium concentration on the grain boundary itself) is likely to influence the occurrence and rate of intergranular corrosion.

Figure 37 provides data from two AGR reactors showing depth of attack vs. cladding irradiation temperature from fuel having achieved different burnup levels [62]. As mentioned earlier, depth and width of chromium-depleted grain boundaries are expected to stabilise with burnup, possibly after ~20 GWd/t U.

The problem caused by in-reactor IGA initiation, during spent AGR fuel wet storage, has been inhibited by caustic dosing to a pH of 11.4. Inhibitor levels are a function of both chloride ion content and pool water temperature; a pH of 11.4 represents an optimised level for pool storage conditions at Sellafield. Although the long-term use of sodium hydroxide as an inhibitor was originally thought to have implications, through dissolution of the protective oxide film on the steel with a resultant increase in general waterside corrosion, recent corrosion studies have not seen any evidence of accelerated general corrosion.

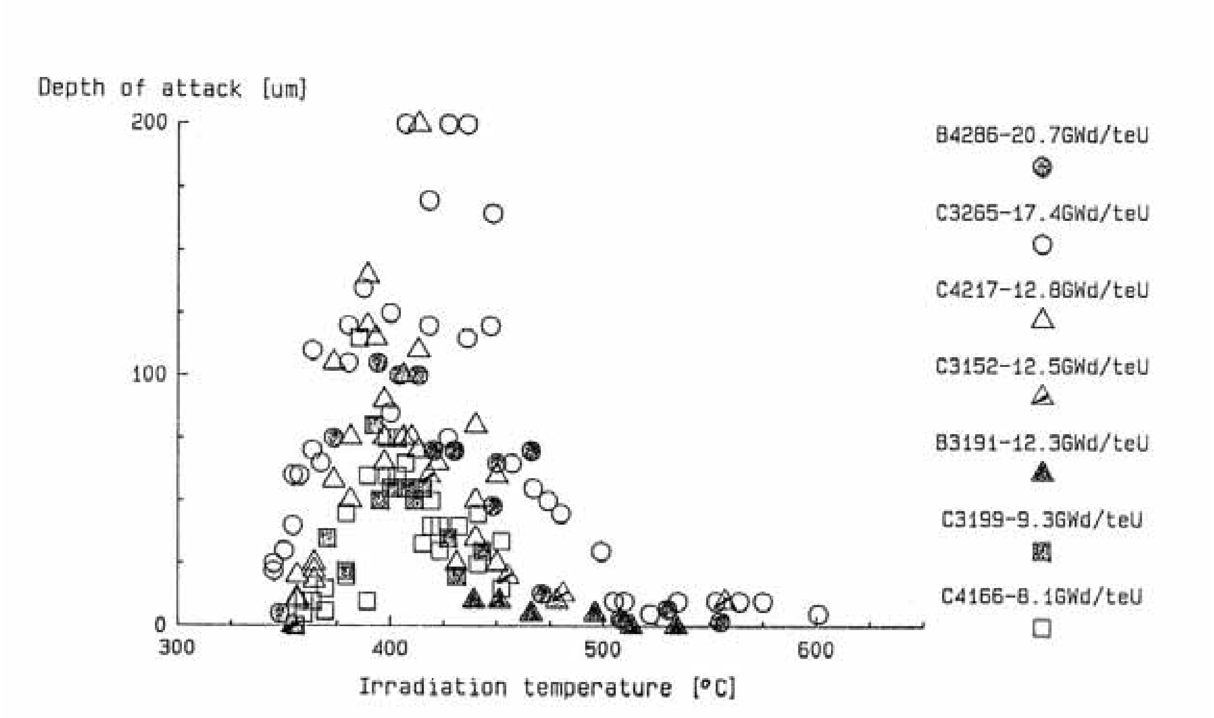


Fig. 37. Stringer-to-stringer variation in mid-wall attack after 72-hour strauss testing (sample exposed to boiling sulphuric acid in contact with copper powder for 72 hour).

7.2.3. Magnox

- Wet Storage

In wet storage MAGNOX-cladding corrodes according to the reaction with water, forming $Mg(OH)_2$ and hydrogen. In principle if the pH is increased to greater than 11.5, the solubility of the magnesium hydroxide layer is low, effectively forming a passive surface film on the metal. Research work, however, shows that this is not the full story and other factors, such as micro-cracking and the presence of carbon in the oxide layer, result in corrosion rates being up to three times higher than unirradiated material. Cladding corrosion is also influenced by the presence of water impurities, notably chloride, which leads to pitted areas even at low concentrations. To minimise these effects chloride and sulphate ion levels are maintained at levels <1 ppm in pond purge water. Pitting corrosion may cause failure of the cladding during longer storage. Crevice corrosion between the splitter blades and the cladding might also occur. For the reasons outlined above, Magnox spent fuel is usually reprocessed within two years of reactor discharge, however, current storage practices show good performance and storage periods >5 years should be expected.

- Dry storage

Since the cladding was developed for CO_2-cooling during reactor operation, dry storage under CO_2 is suitable for long term storage. Corrosion rates in CO_2 are negligible for temperatures below 350°C. Storage in air is feasible when the fuel temperature is below 150°C; the safety case contains restrictions due uranium hydride ignition if bare metal was present. In the case of dry Magnox fuel stored in dry air, fuel deterioration will occur through pitting attack, if relative humidity is >50%, or there is water ingress.

7.2.4. CANDU fuel degradation under dry storage conditions

- Cladding degradation mechanisms

The potential mechanisms of concern for degradation of CANDU fuel, during long-term dry storage, are principally related to hydrogen effects. Typically, spent CANDU fuel has much lower burnup than LWR fuels; this results in lower fission product inventories and lower residual heat, which result in lower storage temperatures. The effect of the lower temperatures is beneficial for most cladding degradation processes, except for some phenomena associated with hydrogen effects. Hydride precipitation and thermal cycling effects are enhanced in a temperature range close to the ductile-brittle transition, which is expected to prevail from the start of the dry storage period.

For fuel elements with cladding defects developed during reactor operation, the potential exposure to oxygen, under abnormal storage conditions, could lead to cladding rupture as a result of fuel pellet oxidation and expansion. This is, however, a process that would occur only under abnormal conditions and would affect an extremely small fraction of the fuel, and, as a consequence, will not be examined here. The possibility of cladding failure by creep rupture has been extensively examined [10] and summarized in IAEA-TECDOC-1243 [4]. Given the low storage temperatures (<200°C) and cladding hoop stresses (<100 MPa), creep rupture is not considered to be a limiting mechanism for CANDU fuels. For CANDU fuel, Delayed Hydride Cracking (DHC) of the endcap/endplate welds is considered to be the most critical mechanism that could affect the bundle integrity during dry storage.

- Delayed hydride cracking (DHC) test methodology and test apparatus

To further gather information on the properties of the bundle endcap/endplate welds and to determine whether Delayed Hydride Cracking (DHC) could be a concern, an apparatus and methodology were developed that could be easily implemented in a hot-cell to test endcap/endplate welds of irradiated fuel. Parameters of interest include the stress intensity factor that can lead to endcap/endplate crack formation and a determination of the crack velocity of endcap/endplate welds of irradiated fuel as a function of temperature in the range of -50°C and 150°C relevant to dry storage conditions.

In 2007, the first phase of the program was completed by developing the apparatus and testing the methodology on non-irradiated endcap/endplate welds from a commercial CANDU fuel bundle. The methodology is based on a bending test which consists in essentially peeling off the endplate from the endcap by applying a torque on the weld. The loading supplied by the torque is measured and the growth of the crack is followed by measuring the potential drop along the crack and by acoustic emission. A photograph of the DHC Test Apparatus is shown in Fig. 38.

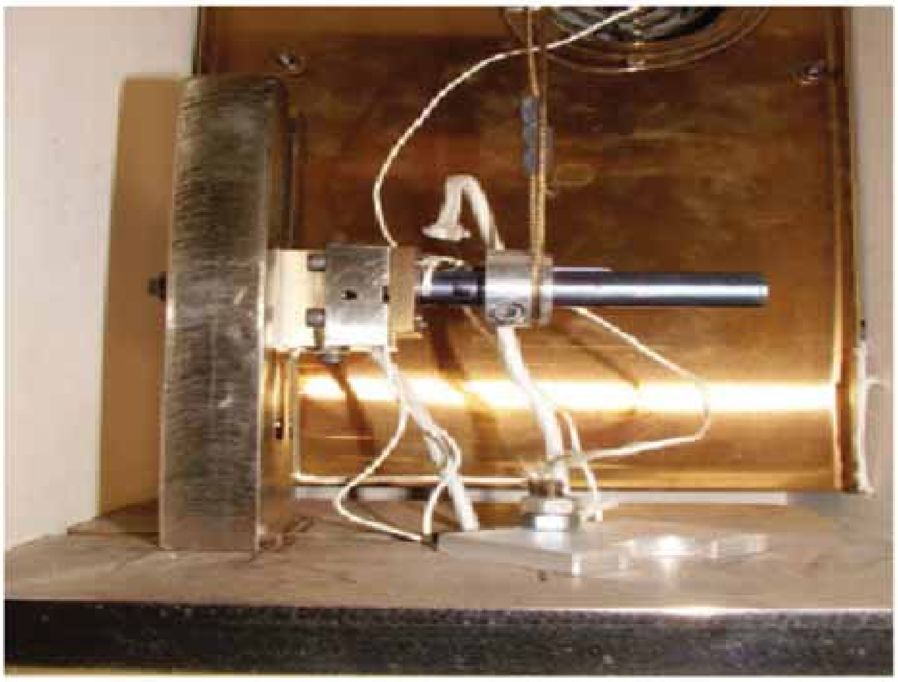


Fig. 38. Photograph showing an endplate/endcap sample in the loading rig being loaded by the cable.

Results of the tests indicated that the test apparatus was successful in initiating and following crack growth of the endcap/endplate weld. A typical crack is shown in Fig. 39.

The crack begins at the notch of the endcap/endplate weld, as indicated in Fig. 39, before proceeding into the weld material in the direction of the endplate as loading from the bending stress is increased. The weld notch acts as a stress raiser from where the crack grows.

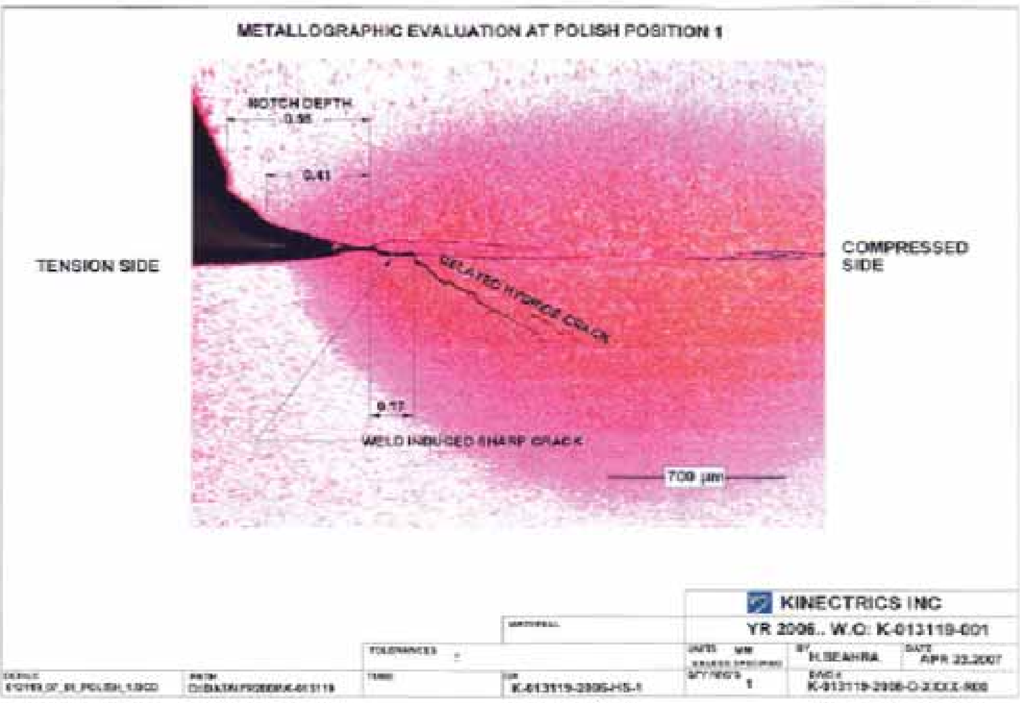


Fig. 39. Metallographic section showing the weld notch, weld discontinuity and DHC crack.

The first test was used to commission the apparatus and demonstrate the methodology. The test material from the fuel bundle has concentrations of hydrogen in the Zircaloy matrix of about 10 ppm and was used to confirm whether DHC could be operative at concentrations of hydrogen which are nominal for a manufacturing bundle, and, also, to scope a series of tests with endcap/ endplate welds with concentrations relevant to those found in irradiated fuel.

The results confirmed that DHC could be operative at this small concentration for a stress intensity factor of 13.9 MPa $m^{1/2}$ at 130°C. In preparation for future testing of irradiated endcap/endplate welds, a series of tests were done at various temperatures and loadings to further develop the methodology and provide benchmarking results.

For these tests, the unirradiated bundle cladding and endcap/endplate welds were loaded electrochemically with hydrogen to a level of about 40 ppm. The tests results were assessed with both analytical stress intensity factor equations relevant to the geometry of the test samples and with a finite element code developed for the purpose. The finite element code calculated stress intensity factors that are in agreement with the analytical models. Delayed hydride cracking was confirmed through analysis of the crack morphology through Scanning Electron Microscopy (SEM). Stress intensity factors (K_{IH}) from the tests are about 7.6 - 13.6 MPA $m^{1/2}$ at 150°C and crack velocities of the order of 10^{-9} m/s.

7.2.5. RBMK and WWER-440/1000

For RBMK and WWER-440/1000 fuel, the assessment of the cladding degradation under dry storage conditions has to focus on the creep behaviour of the Zr1Nb cladding used in all reactor types. A storage programme was carried out to demonstrate the performance of Zr1Nb clad WWER-440 fuel in the temperature range of 330-380ºC. The post-test investigation showed no alteration of the fuel rods, especially no diameter increase beyond the detection limit. These results had been expected, and are in accordance with test predictions. It was concluded that, since the WWER spent fuel with a burnup of 36 GWd/t U exhibited no detectable hoop strain at dry storage between 380 and 400ºC, this experiment proved that WWER spent fuel assemblies behave like other fuels with comparable end-of-life conditions.

7.3. TRANSITION MODES BETWEEN DIFFERENT STORAGE CONDITIONS

7.3.1. Transition from wet to dry storage

In most cases, wet storage of spent nuclear fuel is followed by dry storage. The transition from wet to dry may occur either relatively early (e.g., 3–10 years), if the back end strategy includes dry interim storage, or much later if the spent fuel management strategy includes extended wet storage (e.g., Sweden). Since the mechanical properties of the cladding and structural materials are expected to change very little during the wet storage period, the time of transition will not have a significant influence on fuel performance during the transition process.

The drying process of spent fuel is a very important step during preparation leading to dry storage. Removal of residual water from the storage container and the fuel typically relies on a vacuum drying process that results in increases in spent fuel temperature; this will usually exceed the equilibrium storage temperature, due to the absence of heat transport by the cover gas. For most of the fuel, the stresses and strain, related to the changes in temperature during drying, have negligible effect on the fuel assembly integrity. However, the temperatures reached during vacuum drying could be quite elevated. If the vacuum drying period is relatively short, the temperature increase is limited to a range between 10°C and 30°C. However, since the temperature dependence of hydrogen solubility in Zircaloys is of the Arrhenius type, a significant amount of hydrides may dissolve, and subsequently re-precipitate and partially re-orient upon cooling. Therefore, especially for high-burnup PWR spent fuel with potentially high cladding hoop stresses, temperature limits during drying have to be kept sufficiently low to minimize the potential for hydride re-orientation. Another aspect resulting from the increased temperatures during vacuum drying is the recovery of the irradiation hardening from the cladding material. The recovery starts at about 350°C, and may be considerable for temperatures above 400°C. However, in most of the cases, the recovery has no influence on the estimated dry storage performance.

Fuel handling procedures are normally designed and performed in accordance with the mechanical properties of fuel assemblies. Therefore, based on the above considerations, under normal operating conditions, these operations are such that possible failure modes are unlikely.

In summary, if the handling procedures (designed in accordance with the fuel assembly mechanical design) are strictly followed, no mechanisms exist that might cause the failure of operationally intact fuel elements during a transition from wet-to-dry.

The transition performance of fuel assemblies with **cladding defects**, originating from reactor operation, is however, heavily influenced by the process of water removal from fuel rod internal void volumes. Drying of spent fuel with cladding defects needs a careful adaptation of the drying process to the water release mechanism, to ensure that water is reliably extracted to the desired levels (See Chapter 8). At the increased dry storage temperature, residual water remaining in the fuel rod internal void volume could conceivably react with the UO_2 to form U_3O_8. This in turn could impose mechanical stress on the cladding, due to the resulting volumetric increase with respect to UO_2. Therefore, careful drying is recommended when operationally defected fuel is entering the dry storage.

7.3.2. Dry-to-wet transition

The most critical part of the dry-to-wet transition process is the flooding of the fuel with water. The temperature drop can be sufficiently fast (quenching) to result in high thermal stresses in the cooling zone. However, simple calculations indicate that the stresses remain well below the yield material strength. In addition, the available operating experience shows that the cooling-down process has no detrimental effect on fuel assembly integrity. During dry storage demonstration tests [63], casks were flooded and the cask temperatures were carefully recorded. The total cooling time varied from 1 to 5 hours. The fuel rod quenching at the waterfront could be clearly observed. Flooding of the cask during these tests did not affect the fuel assembly integrity. Intentionally failed fuel was also placed in the test casks. Although a significant increase of the $^{134/137}$Cs activity was observed, there was no further deterioration of the identified cladding defects.

It was concluded from these dry-to-wet transition tests that no mechanisms existed that might cause the failure of intact fuel elements.

8. DRYING

8.1. PURPOSE & BACKGROUND

The loading process starts when storage or transport casks are submerged in the cask pit (or directly into the spent fuel storage pool) and fuel assemblies are transferred from their storage locations in the pool racks into their specific positions in the cask basket. After placing the cask internal lid, the loaded casks are removed from the pool and the different preparation processes start to transfer the fuel assemblies from a ‘wet’ to a ‘dry’ inert environment.

Drying is considered to be composed of 2 main stages:

- the removal or draining of the bulk amount of water contained in the cask cavity, and
- the elimination of residual amounts of water and vapour contained in the cavity and/or inside the fuel assemblies (in case of damaged fuel)

The drying process is of a paramount importance to the dry storage systems as it prevents:

- the presence of any oxidizing gases (CO_2, O_2 and others which may be formed by radiolysis of residual amounts of water) during the dry storage period, thereby guaranteeing that fuel assemblies will be stored in an inert, non-corrosive environment and that these gases will not cause an unacceptable pressurization of the cask cavity.
- The presence of hydrogen from radiolysis and the potential buildup of flammable gas mixtures in the cavity of the packaging during the storage period.

8.2. TECHNIQUES

Draining is usually accomplished either by using a hydraulic pump, by blowing gas (air, nitrogen, helium or argon) into the cask cavity through one opening which forces water to exit through a second one, or by mere gravity (in those casks with a penetration in its bottom part).

Drying consists in removing the residual water and water vapour present inside the cask. The drying process is somehow more complicated and time consuming than draining. It is normally demonstrated by achieving a certain low pressure inside the cask cavity and being able to maintain it throughout a fixed period of time or by measurement of relative humidity of the remaining gas. Two main techniques are nowadays employed and can be combined: vacuum drying and hot gas recirculation, which are hereinafter briefly explained.

8.2.1. Vacuum drying

The theory of operation is simple: once the cask lid is in place and tightened in an appropriate way to create a sealed environment and the cavity drained, the cask internal pressure is lowered by connecting a vacuum pump through a cask cavity opening. As the pressure goes below the saturation pressure for the temperature of the water inside the cavity, the water boils and the vapour is carried out of the cask by the vacuum pump.

Water evaporates rapidly under vacuum conditions and the time it will take the water vapour to be removed from the cask depends on a variety of factors such as the dimensions of the spaces which the vapour has to flow through (port openings, clad cracks in case water is trapped within the fuel assemblies, specifics of the design of the cask internals, etc.).

Spent fuel assemblies residual heat aids in the whole vacuum drying process. Insufficient heat (i.e., low ambient temperatures or very long cooled fuel assemblies) may make difficult the drying task. It is recommended to have a minimum gas temperature of 10 °C in the cask to prevent condensation of humidity. In case of freezing water, impracticably longer drying respectively detection times are necessary due to the very low vapour pressure of ice. To prevent that, there are several possibilities which can be implemented in the drying procedure: to heat the cask before putting it into vacuum, or to maintain the pressure above 6 mbar which is the pressure of aqueous vapour over water for a temperature of 0°C.

8.2.2. Hot gas recirculation

The principle consists in forcing a continuous circulation of helium (or another inert gas) through a set of modules which basically heat the cask internals to aid removing the moisture from the cask cavity. The helium-moisture mixture is transferred to condensers where the moisture is removed from the helium stream. This moisture removal process may be further enhanced, once the initial condensing is accomplished, by freezing the remaining moisture out of the flowing helium stream, in order to achieve the required dryness in the cask cavity.

As the cask internals are permanently under a helium environment, which is the main condition under which the cask thermal analyses are performed and evaluated, there are no time limits associated to the use of this technology, which may be of particular importance in case of drying a high heat capacity cask or a set of severely damaged fuel assemblies.

8.3. SOURCES OF RESIDUAL WATER IN THE CASK CAVITY

Water may be present in the cask cavity under a variety of forms:

- As free water
- Absorbed (either physisorbed or chemisorbed) water in the fuel itself and all structural components
- Chemically bounded water (crud deposits, other corrosion products and boric acid)

Free water is normally easy to be removed by using one or a combination of the techniques identified above. Physisorbed water molecules are normally found in all surfaces of the fuel assemblies and cask internals and are not too difficult to remove as their binding energies are generally low (few kcal/mole) if sufficient temperature is provided.

Chemisorbed water molecules present binding energies which are generally one order of magnitude higher than physisorbed ones and they are more difficult to extract. Different compounds may be formed as a result of water reactions with the fuel pellets (due to cladding failures) and water corrosion of the cladding and/or cask materials. The chemical forms comprise hydrates of different uranium oxides and several metal hydroxyls and oxyhydroxides. When these chemical forms are susceptible to release water at the normal storage temperatures, they need to be preliminary dehydrated which can be achieved at higher temperatures, depending of pressure. This is not necessary when the dehydratation occurs at temperatures which are higher than design storage temperatures.

Finally, water may be contained in the crud deposits on the fuel assembly surfaces or in other hydrated corrosion products, although they are typically present in negligible quantities.

8.4. DRYNESS CRITERIA

Countries as the US have established criteria which recommend limiting the maximum quantity of oxidizing gasses to a total of 1 gram-mole per cask, which corresponds to a concentration of 0.25 volume % of the total gases for a 7.0-m^3 cask gas volume at a pressure of about 0.15 MPa (1.5 atm) at 300ºK [64].

In practical terms, this criterion is considered to be fulfilled, considering that the drying operation was carried out at a pressure always higher than 4 torr, if a pressure around 3 or 4 torr is achieved in the cask and maintained over a period of about 30 minutes, once the cavity has been isolated from the drying system.

In Germany, an early dryness criterion was based on the corrosion of the primary lid sealing. Stipulating a 20% wall thickness reduction, 270 g of water could be allowed per m^2 of the visible sealing area equivalent to 3-5 g water in the atmosphere per m^3 free volume for typical casks. The residual water in the cask can be measured by determination of the dew point of the evacuated water vapour.

Today, a high corrosion resistant sealing is used and the timely behaviour of the residual humidity in the cask considered. Using such an integral methodology, for instance for a CASTOR cask, the dryness criteria can be linked to a vapour rate of 0.7 hPa per hour. Such a rate is equivalent to a water content of about 120 g for a typical cask. Nowadays, the pressure rise criterion for dryness after vacuum drying is more and more used.

For the hot gas recirculation method, the procedure normally used controls the temperature, pressure and recirculation rate of the gas in the cask cavity throughout the different stages of the process. The dryness criteria is fulfilled if the gas temperature at the outlet of the moisture removal component of the system or the dew point of the gas exiting the cask cavity is below a certain temperature for at least 30 minutes.

8.5. ISSUES

- **Fuel cladding temperature limits**

  A major issue (inherent to vacuum drying) is related to the poorer heat dissipation capacity of the fuel assemblies within the cask under vacuum or near to vacuum conditions (as compared to a helium filled cask) for long periods of time, which may subject fuel cladding to higher temperatures than the ones recommended for short-term operations.

- **Icing**

  During vacuum drying, as water boils inside the cask, the cask internal temperatures may decrease. Localized cooling may occur within the cask, making it possible to freeze the residual water, which would make the water removal process very slow or would possibly stop the water removal.

  In normal practice, due to the decay heat from the fuel (in standard commercial light water reactor fuel assemblies), icing will be more likely to occur in the evacuation hoses than in the cavity itself. This effect may cause clogging of the lines or system connections.

  If pressure is maintained above 6 mbars (4 torrs), the risk of icing is avoided.

- **Insufficient heat from the fuel assemblies**
  When loading fuel assemblies with extremely low heat loads, the vacuum drying process may be very time consuming and experiences have been reported in which an external heat source has been applied to improve performance.

- **Damaged fuel assemblies and fuel debris**
  In those cases where it is suspected that there are major quantities of chemisorbed water (in severely damaged fuel assemblies or non-standard fuel elements) which may be quite difficult to remove from the cask, it may be advisable to purge the cask or to re-vacuum-dry it periodically.

  In some countries other options were applied:

  In France (TN International), recombiners of radiolysis gases have been qualified.

  In Germany, the regulation usually requires canning of damaged fuel. A use of drying agents is a special measure to handle fuel whose cladding damage cannot be clearly excluded.

8.6. EXPERIENCES

(1) Canada

- Drying

In Canada, both vacuum drying and blown air drying are being used depending on the Dry Storage System. In non-transportable systems such as AECL’s CANSTOR and Concrete Canisters, fuel is first retrieved from the waterpool in a special flask before being dried by blowing hot air. Transportable dry storage systems such as Ontario Power Generation Inc. (OPG) Dry Storage Containers (DSCs) are wet loaded in the waterpool, and, after draining the pool water, the DSC is transported to a Dry Storage Processing facility for dry vacuuming and sealing before being moved to storage.

- Drying with Heated Air in the CANSTOR System

The CANSTOR system is a convection air cooled modular reinforced concrete structure with capacity for 12 000 CANDU bundles. The fuel is encapsulated in stainless steel cylindrical baskets or canisters.

A special flask with a shielded bottom door is used to transfer the baskets to the waterpool for loading with fuel. The flask remains outside the waterpool at all times but it is positioned on top of a shielding skirt located in the waterpool to provide shielding when the basket loaded with fuel is returned to the flask. Then, the flask transports the loaded basket to a specially designed shielded workstation for drying with blown hot air. When the fuel is dried, the basket is seal welded with a cover, and, transported to the CANSTOR facility for storage.

The age of the stored fuel is typically at least 6-7 years old with a residual heat of 6.1 watts per bundle. The maximum fuel temperature of the stored fuel is kept below 160°C. The CANDU fuel bundles are stored in the baskets vertically by resting on their endplates.

Vacuum Drying and Helium Backfilling in OPG’s Dry Storage Containers

A Vacuum Drying and Helium Backfilled process is used for the Ontario Power Generation Inc. designed Dry Storage Container (DSC). As transportable units, the DSCs are wet loaded in the nuclear station waterpool. Fuel is stored inside the DSCs horizontally in storage modules at temperatures similar to those of the CANSTOR system.

The Drying process for the DSCs is a two step process consisting of preliminary drying at the water pool and dry-vacuuming at the Dry Storage Processing facility.

- Preliminary Drying at the water pool

An empty DSC is brought to the nuclear station water pool and its base submerged into the water pool for loading with fuel. When loaded, the DSC lid is replaced and secured to the DSC with ‘In-bay’ clamps. The DSC is then lifted out of the water pool with the water in the DSC draining out into the water pool.

When the water pool water has drained, the DSC is moved by an overhead crane to a designated area near the water pool to be washed down for decontamination of its outside walls, and, drained further if required. Following this first drying, the In-bay clamps are replaced with a different set of transfer clamps, and, the vent and drain ports are closed off with specially designed plugs. A vacuum is then applied to remove any remaining water, and, also, to bring the DSC cavity to subatmospheric pressure to prevent egress of radioactive contents out of the DSC cavity during transportation and until the next drying stage. This is accomplished by means of a portable vacuum pump system provided during transportation.

- Dry Vacuuming at the Dry Storage Processing Facility

Upon arrival at the Processing Facility, the DSC lid and the permanent vent port plug are welded before vacuum drying. Vacuuming normally takes about two hours. The DSC cavity interior dryness is regulated by bringing the cavity pressure to below 1 mbarA for at least 30 minutes. The dryness of the DSC cavity can be verified by monitoring the cavity pressure for 15 minutes. If no pressure increase is observed over a 15 minute period, the interior is considered to be dry.

- Helium Backfilling

After vacuuming, the DSC is backfilled with helium through its drain port to a subatmospheric pressure of 930 mbarA (13.5 psiA). If this pressure is exceeded, the vacuum pump(s) can be used to trim the helium backfilled pressure.

The DSC is then leak tested in a Vacuum Box by bringing the pressure in the box to about 5 x 10^{-2} mbars. Leak tightness is guaranteed if no helium is detected.

(2) Germany

Originally, the humidity in CASTOR casks was determined by the dew point mirror method. However, the mirror was often polluted and needed to be recalibrated. This led to widely varying measuring times and the work could not be reliably planned. Due to the change to the pressure-rise method, those uncertainties could be overcome. Dewatering, drying and humidity measurement can now be performed within a very stable time period of 45 hours [65].

(3) Hungary

Fuel is transported from the plant to the MVDS in water filled casks. Before loading in the MVDS, fuel is lifted out from the cask, and part of the water flows back in the cask volume. It is assumed conservatively in the calculations, that there is 0.5dm^3 water on the zircomium surface and entrapped in the crevices. Drying is accomplished in a shielded tube, where hot air (process temperature 90 - 100ºC) flows from the top down and evaporates the water, in addition to blowing it away. This ensures complete removal of residual water from all parts of the fuel assembly. The process time (approx. 1 hour) depends upon the heat input and flow rate of the hot air. The fuel assembly container is considered to be dry, when the moisture content of the exhaust air equals the moisture content of the inlet air.

The same system can be used, when removing fuel from the MVDS: before loading an assembly in the water filled cask, it can be cooled down, operating the same system without heating the air.

(4) Japan

In Japan, the vacuum drying process is applied to dry spent fuel assemblies loaded into the casks. Typical drying process is shown as follows [66]: The cask is drained by the pressure of station service air which is controlled at 0.4 MPa or less. A two step-vacuum drying process is used to dry storage casks. The first vacuum drying is nearing completion in 6 hours. Then, helium gas is charged into the cask cavity and kept for 16 hours to make the cask temperature uniform before the second vacuum drying is performed. Following the helium gas charge, the second vacuum drying is carried out for 1~3 hours. The completion of vacuum drying is judged on a cavity pressure and its pressure rise rate after stopping vacuuming. After the completion of vacuum drying, the cask cavity is filled with helium gas whose pressure is adjusted to the specified initial cask cavity pressure (0.04MPa). In this operation, the amount of cask cavity moisture is checked to be 10 wt.% or less by measuring a dew-point temperature.

(5) Spain

Vacuum drying is used to dry storage casks at the Trillo NPP and the procedure has been successfully applied for all the 14 casks stored up until 2008. Drying times employed to fulfil with the approved criteria has been less than 30 hours in all cases.

Hot gas recirculation will be used in the near future to dry store all the fuel assemblies coming from the Jose Cabrera reactor. A population of 23 damaged fuel assemblies will be dried using this method. Loading operations are schedules to start early in 2009.

(6) Sweden

- Drying

The spent fuel from the power plants is transported to the interim storage for spent nuclear fuel, CLAB, in dry transport casks. The fuel is dried in the transport cask by putting the cask under vacuum. The ‘dryness’ is determined by checking the pressure rise in the cask during a given time.

The surface of the cask basket was a cause of drying problem because of a porous structure that kept water that was hard to evaporate by vacuuming. The baskets with this porous surface have been changed.

- Leaking fuel (transport of spent nuclear fuel from NPP to interim storage, CLAB).

Procedure at the NPP

1 Water sample from fuel pond, ^{137}Cs
2 The transport cask is loaded with the spent fuel
3 After 4 hours a sample is taken from the cask and analysed for ^{137}Cs
4 If the ^{137}Cs concentration is raised by more than 5 E4 Bq/kg the fuel is considered as leaking.

- Experience of transporting of leaking fuel

Transport of minor leaking fuel from NPPs to CLAB has been executed. The experience is good and no further damage to the fuel because of the transport and cooling process at the reception at CLAB was observed. Prior to the transport, the fuel assembly is inserted into a special box that is open in the bottom and in the top by valves that also contain filters. The purpose of the box is to prevent any possible fuel fragments/debris that can come loose from the fuel to contaminate the transport cask and the cooling system at CLAB. When the fuel has been unloaded at CLAB the fuel assembly will be taken out of the box and it will be treated as an intact fuel assembly under the condition that the fuel does not have any major damage as broken rods or similar damage.

(7) United Kingdom

- Drying AGR Fuel

The drying of AGR elements was investigated as part of the development programme for the proposed Scottish Nuclear Limited (SNL) MVDS in the 1990s [67]. Unlike other fuel clad materials the drying requirements for AGR fuel clad (Nb stabilised 20Cr-25Ni stainless steel) are more onerous due to the clad susceptibility to stress corrosion cracking (SSC). SSC in this case is initiated through a build-up of nitric acid in the pores of the clad microstructure. Nitric acid is formed from radiolysis of residual moisture in the presence of nitrogen impurities.

To meet the target moisture content of the dry storage tubes cover gas (<50 vpm) a combination of forced hot air (180ºC) drying in flowing air (for drying the fuel pins) and induction drying of the graphite sleeve (to remove tightly bound water) was shown to be effective. Selective drying of the graphite sleeve was chosen to over-come the safety issues associated with UO_2 to U_3O_8 conversion; if any failed fuel was present.

The drying process is specific to AGR whole elements and would have been incorporated as a dedicated facility within the proposed MVDS. It is unknown whether the whole or parts of this process could be applied to other dry store technologies for AGR fuel storage.

8.7. RECOMMENDATIONS

For **draining**, in those cases with a drain line inside the cask cavity, a small amount of water in liquid phase may still remain inside the cask because the drain line ends above the cask bottom. This remaining water may be minimized by tipping the cask so that the drain line is at the low point.

For **drying**:

Several lessons learned from known practices (mainly with the vacuum drying technology) include the following actions:

- Test the vacuum lines and overall system integrity prior to cask loading (the cask cavity may be dry but vacuum may not be maintained due to losses throughout the lines or connections);
- Minimize the number of connections and lengths of the lines to reduce the possibility of inleakages; optimize the cross section of the lines in dependence of the vacuum pump performance.
- It is convenient some times to break vacuum conditions (by circulating nitrogen, helium or air) several times to aid for water vaporization and prevent ice formation; or ensure sufficient heat transfer by continuous helium mixing while drying of the cask.
- Use a vacuum skid with a redundant vacuum pump or have a spare pump available to avoid having to unload the fuel from the cask in case of a malfunction;
- Measure the quantity of water extracted from the cask cavity in every stage of the preparation process;
- Measure of the dew point of exhaust gas, representative of the dew point inside the cavity.
- Document all parameters and readings throughout the vacuum drying process.
- Tightening of the sealing in an appropriate way means to dry the whole surface of the sealing before final closing of the lid.

9. BEHAVIOUR OF STORAGE FACILITY COMPONENTS

The behaviour of all materials used in spent fuel storage facilities has been extensively reviewed [68] and IAEA Safety Series No.116 and No.117 provides guidance on design and operation of spent fuel storage facility [69].

In the case of the internal environment of dry storage systems, excluding air filled systems, the only conditions, which exist for corrosion, are from residual moisture and pool chemical additives remaining after loading operations. All external surfaces and interfaces, however, are still susceptible to attack.

In the case of wet storage all materials corrode in the storage environment, the variance is in the rate of corrosion of the different materials. For example, unprotected mild steel has a significant corrosion rate in water. The second point to remember is that as storage duration increases up to 50–100 years, the general corrosion rate for some materials used in storage systems can become a life-limiting factor. Thirdly, additives, which have been used to minimize other corrosion mechanisms or for system operability purposes, may have detrimental effects on the long-term use of some storage components.

In the following chapter only materials where there has been active research and development by participating SPAR II countries will be reported. New areas of interest include the development of neutron shielding and criticality control materials for application in transportation and storage casks, and the continued development of reinforced concrete dry cask systems.

9.1. REINFORCED CONCRETE

Concrete is by nature a non-uniform non-homogeneous material. It is relatively inexpensive and, though it has high compressive strength, is weak in tension. Steel, which is more expensive, has very high tensile strength, is therefore designed to be encast into concrete in regions of tensile stress thus taking over from the concrete in carrying these tensile effects. Steel also has the beneficial effect on concrete of controlling the width of cracks associated with tensile strains in concrete. Such tensile strains may be caused by mechanical and thermal loading, and/or the effects of restrained drying out shrinkage (soon after casting), as well as surface thermal contraction strains due to temperature gradients during curing conditions.

Good quality modern concrete in a benign environment can be expected to last indefinitely without any serious deterioration in structural properties. Conversely, unlike plain concrete, reinforced concrete is generally not as inherently durable, owing to the propensity of its steel reinforcement to corrode. ‘Cover’ is the specified minimum thickness of concrete from the reinforcement to the external concrete surface. Once the concrete in this cover zone to the reinforcement looses its alkalinity, and given that moisture and oxygen can get to the reinforcing steel, the reinforcing steel will start to corrode.

There are many possible causes of degradation in reinforced concrete structures; they include (i) physical conditions such as movement of the foundations, structural over loading, accidental impact damage, (ii) thermal cycling, and (iii) chemical/environmental effects; for example Carbonation or Chloride ingress.

It is, therefore, imperative that concrete structures are regularly inspected (this is usually done on annual basis for large wet storage pools) so that warning signs can be detected and the causes of defects identified.

Current R&D interests have mainly been focused on the use of reinforced concrete in cask systems for the dry storage of LWR fuel. The main factor which differentiates its application in dry storage c.f. wet is the operating temperature can be up to 90ºC in the early years. At these higher operating temperatures the durability of the reinforced concrete can be affected considerably; for example it has been shown that the depth of carbonation at 65ºC is twice that of concrete operating at 20ºC after 5 years [4].

9.1.1. Canada – changes in cement used in dry storage container (DSC) manufacture

Due to concerns with the long term availability of Canadian Standards Association (CSA) Type 20 cement (i.e., moderate heat of hydration), two main studies were conducted to evaluate the viability of replacing the CSA Type 20 cement, which was being used in the manufacture of the DSCs, with:

- CSA Type 20E or Type 50E equivalent cements. In particular, replacing about 20% Ordinary Portland cement (OPC) with Ground Granulated Blast Furnace Slag cement (GGBFS or 'slag').

- CSA Type 50 cement (i.e., high sulphate resistant cement).

Regarding slag cement, a technical review of its properties and its potential substitution of Type 20 cement was completed. No data was found in the literature on high-density slag cement blended concrete. However, based on the review of available data on slag cement used in normal density concrete, it was concluded that slag–based equivalent cements hold promise for satisfactory use in the DSCs. The review cautioned, however, that further work was needed to investigate a concern with the effect that the sulphide content in the slag cement could have on corrosion of the C-steel reinforcement in a 'closed system', such as the DSC concrete wall. Also, the review identified that radiolytic effects on slag cement concrete merited further assessment, although it was felt that this was a minor issue that needed further confirmation due to the lack of information.

A follow-up thermodynamic study on the potential for the concentration of sulphide present in slag cement to be converted to hydrogen sulphide in the enclosed system of the DSC concrete wall was performed. The results of that study pointed to the possibility that conversion of the sulphide to hydrogen sulphide under the conditions in the DSC wall might be possible if sufficient residual water remains to form an aqueous phase in the concrete matrix of the DSC wall. If only a water phase vapour remains, then the generation of H2S would be expected to be insignificant. The study pointed out the importance of a low residual moisture content in the slag concrete to reduce the risk of enhanced corrosion of the reinforcement and DSC steel components. Kinetic rate studies to confirm the thermodynamic findings were not undertaken.

A development programme for replacing Type 20 cement with CSA Type 50 in the high-density concrete used in the DSCs was implemented. The programme included multi-phase laboratory testing and a 'field trial'. The purpose of the laboratory tests was to develop, optimize and test a candidate high-density concrete using Type 50 cement. The 'field trial' was designed to confirm full–scale performance of the concrete, as well as to address practical aspects of its processing such as mixing, batching and curing.

Satisfactory results were obtained with the Type 50 cement, and, the experimental findings met the specifications for the DSC, as well as the requirements for long term performance and durability. Type 50 cement met a 7-day heat of hydration limit set at 300 kJ/kg cement, as per the DSC specification. Results from the field trial indicated that some localized honeycombing/layering was observed in the hardened concrete, which was attributed to inadequate compaction during placement, and, was later resolved.

The main conclusion of the developmental programme proved that Type 50 cement can be recommended for use as a replacement for Type 20 cement in producing the DSC high-density concrete.

9.1.2. Japan – concrete cask development programme

Since 1997, the CRIEPI has been conducting research programmes into the interim storage of spent fuel in casks; this has mainly involved concrete cask storage technologies, which applies to making the technical requirements issued by NISA/METI of the Japanese government. Moreover, to verify cask integrity under long-term dry storage conditions the following development work has been undertaken (see section 5.3. Table VIII);

Two types of full-scale concrete cask and two types of multi-purpose canister (MPC) were designed and fabricated for the following demonstration tests:

- Thermal characteristics of the concrete casks
  - Heat removal tests under normal and accident conditions
- Structural integrity of the MPCs
  - Drop tests onto the unyielding target
- Seismic tests of a freestanding full-scale concrete cask
  - To understand the tipping-over characteristics
- Stress corrosion cracking evaluation tests of the MPC

(1) Concrete cask design and fabrication

Two types of concrete casks were designed for verification tests. One is a reinforced concrete cask (RC cask) and the other is a concrete filled steel cask (CFS cask). The RC cask is made from reinforced concrete that acts as both structural component and shielding material. On the other hand, in the CFS cask, the steel shell is the structural component and the concrete only provides radiation shielding material. These concrete casks utilize natural convection for heat removal; where the cooling air flows between the annulus gap between concrete container and canister.

Two types of the canisters which differ in construction and basket material were designed. The basket of Type I consists of guide tubes and stainless steel plates. The basket of Type II consists of rectangular hollow blocks made of aluminum alloy. In this test, Type I was used for the RC cask and Type II was used for the CFS cask. For each canister body, high corrosion-resistant material is used. Each canister has a storage capacity of 21 PWR spent fuel assemblies. Decay heat was simulated by electric heaters.

To perform the heat removal tests and drop tests with full-scale casks a demonstration test facility was constructed in the Akagi Test Center of CRIEPI. In this facility, there are heat removal test area and drop test area. On the other hand, seismic tests using full scale cask were performed using 3D shaking table at the National Research Institute for Earth science and Disaster prevention (NIED).

(2) Heat removal test[71]

Temperatures of each position in the cask and flow rate in the normal condition and the accident condition were measured using two types of the full-scale casks as described above, and the integrity of components of the casks was evaluated.

- Normal condition (with a heat rate of 22.6 kW corresponding to the initial storage stage, and a heat rate of 10k W corresponding to the final storage stage)

In the RC cask a maximum temperature of 91°C was recorded for the initial storage stage condition; which exceeded the long-term temperature limit of 90°C. The reason would be that the pressure loss on the natural convection was too large in the flow channel of the cask. Therefore, the thermal design (ex. flow channel shape) must be changed. The heat removal efficiency of the cask was found to be 80% of the total heat and the heat transfer from the bottom of the concrete container to the floor was insignificant.

In the CFS cask the maximum temperature observed during the initial storage stage condition was below the long-term temperature limit for the concrete container.

In both casks, the surface temperature of the bottom part of canister was below 100°C under final storage stage test conditions. Given the operating temperature is above 50ºC; this may leave the stainless steel canister susceptible to the stress corrosion cracking.

- Accident condition (during initial storage stage with 50% blockage of the air inlet)

Tests were performed for accident conditions: Two of four inlets were partially blocked. In both casks, the maximum temperature observed was below the short-term temperature limit of 175 °C within 24hours (Table 6). The increase of temperature in the concrete container was 5°C in the RC cask and was 10°C in the CFS cask, respectively. Therefore, it was found both casks had enough heat removal capacity under the accident condition.

TABLE 6. TEMPERATURE AND FLOW RATE AT 22.6 KW UNDER THE NORMAL CONDITION AND THE ACCIDENT CONDITION OF 50% BLOCKAGE OF THE INLET

Cask Type / Item	RC cask		CFS cask	
	Normal	50% blockage	Normal	50% blockage
T_{in} of air (˚C)	33		33	
T_{max} of concrete body (˚C)	91 (90 *)	96 (175 **)	83 (90 *)	93 (175 **)
T_{max} of canister surface (˚C)	209	214	192	200
T_{max} of guide tube (˚C)	301	306	228	235
ΔT of air (˚C)	65	70	52	66
Flow rate (kg/s)	0.335	0.321	0.363	0.280

* Allowable Temperature Limit Value for the long-term: 90°C [73]
** Allowable Temperature Limit Value for the short-term: 175°C (within 24 hours) [73]

(3) MPC drop test [74]

Two drop tests in the horizontal and vertical condition were conducted at drop heights of 1 m and 6 m, respectively. The canister contained dummy steel structures equal to the total weights of the spent fuels (14.7 tonnes). Figure 40 shows the vertical drop test from the height of 6 m.

During the horizontal drop test, the test canister was slightly deformed near the impacted area. The time histories of accelerometers and strain gauges at various points in the test canister were measured. The average deceleration value was about 436 g at the top of the lids. On the other hand, in case of the vertical drop test, although the bottom plate of the test canister was deformed by the force of inertia of the contents, the deformation of the bottom of the basket was negligible. The average deceleration value was about 1 153 g at the center of the shell.

Helium leak tests were performed before and after the horizontal and vertical drop tests to confirm leak-tightness (especially the welded lids) as a function of the impact loads. Measured leakage rates for the welded lids and canister shell in both tests were under 1.0x10-9 Pa.m^3/s.

Fig. 40. Canister drop test.

(4) Seismic test

A full-scale concrete cask and a storage facility floor were constructed and placed on a three-dimensional shaking table in 3-D full-scale earthquake testing facility ‘E-Defense’ designed and constructed by NIED as shown in Fig. 41. One full-scale PWR fuel assembly (17 x 17) and 20 dummy PWR fuel assemblies, whose modal deformation in an earthquake was equivalent to the full-scale assembly, were fabricated for the trial. A 80cm thick and 8m wide reinforced concrete slab (weight 125 tonnes) was used as the storage facility floor model. During the seismic excitation test, the angle, angular velocity, acceleration, and displacement of the cask body, canister, and fuel structures were measured. For input of the seismic excitation test, two recorded waves during typical natural earthquake waves and one artificial seismic wave were employed. Test conditions also included cases of horizontal and vertical motions simultaneously.

Test results showed that the tip-over of the full-scale cask did not occur and the maximum deformation of the fuel remained under the elastic region; for interactions between the canister and cask body and the fuel and basket. Special attention, however, should be paid for the sliding or jumping behaviour of the cask under the strong earthquake motion.

Fig. 41. 3D-full-scale earthquake testing with full-scale concrete cask.

9.2. NEUTRON ABSORBERS USED IN STORAGE RACKS, BASKETS ETC. FOR CRITICALITY CONTROL

Common neutron absorber finding use in spent fuel storage racks, transportation baskets etc. as a means of maintaining sub-criticality are listed below:

- Boron carbide in a silicone rubber binder; for example Boraflex.
- Boron carbide dispersed between aluminium alloy cladding; for example Boral.
- Boronated aluminium alloy
- Boronated stainless steel alloy
- Cadmium metal in a leak proof cladding; for example Cadminox

The latest designs for transportation and storage casks has called for new neutron absorbing materials to attenuate the higher enriched (^{235}U 5%) and burn-up fuels (>55 GWd/t U); i.e. a requirement for better thermal and neutron efficiency. More recently interest has been directed towards the use of Metal Matrix Composites (MMC) derived from aluminium alloys to act as both neutron absorber and structural component for spent fuel baskets. Aluminium offers excellent thermal conductivity.

One of two technologies are deployed to incorporate the neutron absorbers:

- Adding boron into molten aluminum (ingot metallurgy);

- Mixing the stable compound such as B_4C with aluminium alloy powders (powder metallurgy).

Boron carbide loadings of up to 40% are claimed [75] and boron carbide mixed with aluminum, magnesium and samarium have been investigated [76] under PWR and BWR storage conditions. Examples of application of these new materials are in the MHI MSF [77] and TN 97L [78] transportation and storage casks.

9.2.1. France – Use of as neutron absorber in transportation and storage casks

TN International has been working with Alcan since 2003 in developing a new basket for its TN24 cask design. A phase in fabrication of the casks is shown in Fig. 42. Alcan uses the ingot metallurgy process to produce aluminium alloy based MMC materials with up to 25 w/o boron carbide.

The materials have been subjected to a qualification programme including neutron absorption, thermal, physical and mechanical tests; reported [78]. The first TN24 cask containing the new MMC basket was loaded in 2007.

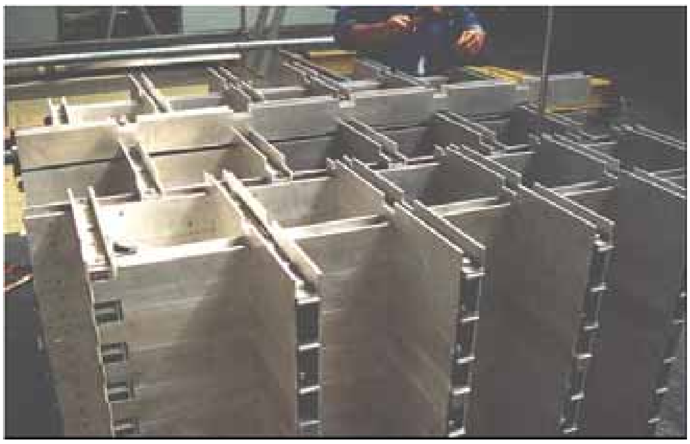


Fig. 42. Fabrication of TN 24 basket.

9.3. NEUTRON SHIELDING MATERIALS

As the name implies the purpose of neutron shielding materials is to stop neutrons. This is accomplished by capturing them with atoms that have a high neutron cross section (normally boron is employed), and in the case of fast neutron slowing them down first so they can be captured; this is achieved by the use of materials with high concentrations of hydrogen atoms (for example polymers).

A variety of polymeric materials have been used as neutron shielding materials these include: Polypropylenes; Polyethylene; Polyesters: Epoxy resins; and a variety of mixed Polymers. The common feature of all polymers is that they degrade with time primarily due to the effects or radiation, and heat. When they degrade they lose weight releasing chemical compounds such as hydrogen chloride, water and oxidised species.

The main requirements of neutron shielding materials to be used in transport/storage casks is their long-term thermal stability and an ability to with-stand high temperatures for short periods of time; i.e. 800ºC for 30 minutes. With a move to increased fuel burn-ups and Mixed Oxide fuels there are increased demands in terms neutron/gamma dose and cask operating temperature which has called for the development of new materials.

9.3.1. France –development of a new neutron shielding material for transport/storage casks

To meet current and projected fuel burn-ups TN International has been evaluating new shielding resins [80] for application in their range of transport/storage casks. High performance shielding materials are required for the severe neutron and gamma source.

The new requirements are to accommodate:

- More efficient (hydrogen and Boron content)
- Fire resisting (M1)
- Resistance to long term high temperatures (160°C to 180°C)

The latest material to be evaluated is Vyal resin which is a composition formed by a reaction between the double bonds of a vinyl-ester polymer with those of styrene to form a highly cross linked structure; source of hydrogen. The mineral fillers alumine hydrate (fire retardant and source of hydrogen) and zinc borate (fire retardant and source of boron) are dispersed within the polymer matrix.

When compared to other neutron shielding materials, polyester resin and ethylene-propylene-rubber (EPR), Vyal resin shows less weight loss at the same temperature for the same sample size and test duration. At 160 ºC Vyal resin lost ~2.5% of its weight after 10 000 hours, this compares to ~3% by EPR and >4% by the existing polyester resins. Figure 43 shows the % release of hydrogen with time for Vyal resin.

Similarly the fire resistance was found to be excellent as the material was found to be self-extinguishing and only lost 7% of its weight.

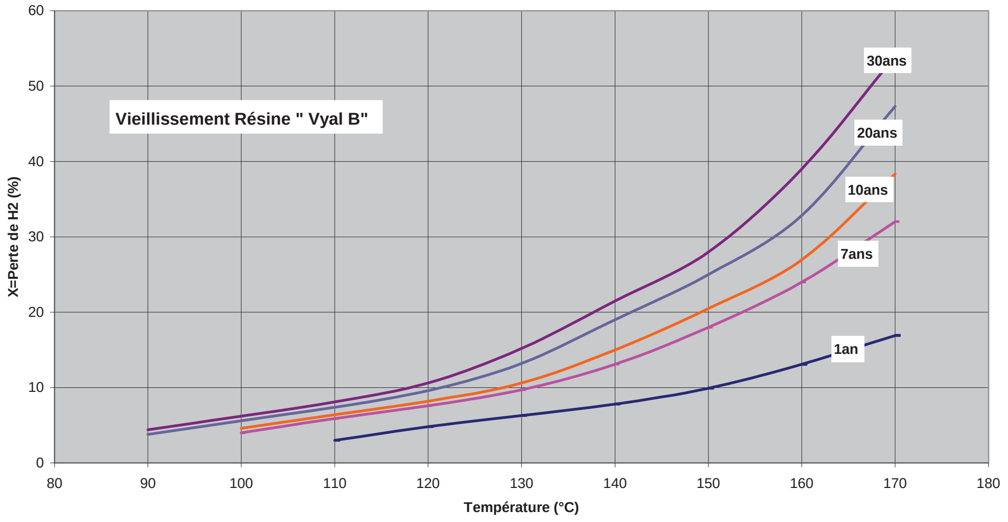


Fig. 43. % release of hydrogen with time.

9.3.2. Japan – degradation of neutron absorbing materials

The Japan Nuclear Energy Safety Organization (JNES) conducted the study of ‘Metal Cask Storage Technology Verification’ up to the end of FY2003. In the study, many tests were conducted to investigate material degradation properties of the main components relating to cask safety [82]. The test results regarding neutron shielding material degradation are summarized.

Epoxy resin, silicon resin, or pyrolitic graphite (PG) can be applied to metal casks as neutron shielding materials. Degradation of the shielding due to thermal aging and irradiation were investigated and a method of predicting shielding performance derived.

For epoxy resin and silicon resin, neutron and gamma ray irradiation tests, heating tests in/ after/ without irradiation were conducted in closed system with forced ventilation, with constant pressure controlled, or without ventilation. The maximum temperature at the beginning of storage was estimated to be 163°C and decreased year on year; decay heat dependent. Degradation was determined for given temperatures, dwell times and cumulative irradiation dose. Temperature of heating was from 130 to 170°C, and the maximum period was 15 000 hours. Cumulative irradiation neutron and gamma ray doses were up to 1.5×10^{15} n/cm^2 and 3.9×10^4 Gy.

After heating or irradiation, weight loss, appearance, chemical composition and gas were determined. The main findings were:

- Weight loss was estimated to occur by release of oxide products of low molecular weight from base materials and of H_2O due to dehydrate reaction of tri-hydrate-alumina.
- The influence of heating on weight loss was dominant, and that of neutron and gamma ray irradiations was small. In addition, there was no synergistic effect of heating and irradiation.
- Relation between weight loss and environment of the tests was: closed system with forced ventilation > with content pressure controlled > without ventilation

Weight loss was selected as an index of the degradation of shielding performance; this was plotted against Larson Miller Parameter (LMP) as shown in Fig. 44 for epoxy resin.

LMP was defined by the following expression:

$$LMP=T\,(C + \log t)$$

T: absolute temperature of heating (K)
C: constant, t: heating time (hour)

As shown in Fig. 44 it was confirmed that a linear relation between weight loss and LMP existed. In combination with knowledge of the operating environment, it is considered that the relationship can be used to give an indication of neutron absorber degradation for epoxy and silicon resins.

In the case of PG water, long term behaviour of the material is influenced by the product of decomposition which raises the freezing point of the material. Neutron and gamma ray irradiation testing, heating tests after/without irradiation, chemical analysis of remaining water and released gas, and determination of the freezing point were carried-out. Degradation was determined for given temperatures, dwell times and cumulative irradiation dose. Specimens were heated from 100° C to 180° C with a maximum dwell time of 10 000 hours, and a cumulative neutron and gamma ray irradiation doses of up to 5.0×10^{14} n/cm^2 and 2.0×10^4 Gy.

The main findings are provided below:

- PG density decreased by a factor of 3% after 10 000 hours at 160°C
- Acetone, aldehyde (precursor of organic acid), oxygen, and hydrogen were detected as major decomposition products. There was little evidence of organic acid.
- A little general corrosion and no SCC occurred on inner surface of casings of SUS.
- Freezing point was only slightly changed, i.e. from -46.4ºC increased to -42.9°C in the test at 160ºC for 10 000 hours.
- The usable limits of environmental conditions that would result in weight loss and thus shielding performance for each of the components are summarized in Table 7.

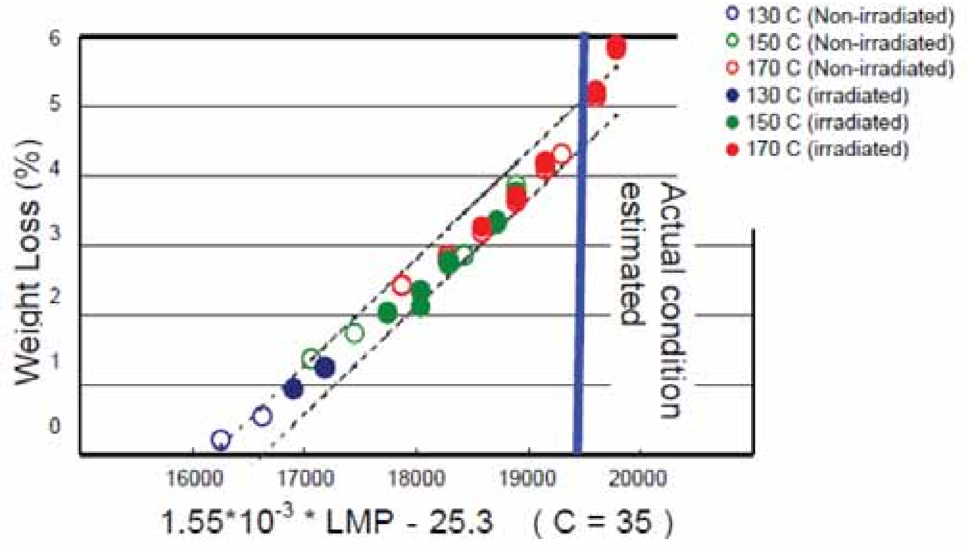


Fig. 44. Plot of weight loss versus LMP for epoxy resin.

TABLE 7. USABLE LIMITS FOR EACH OF THE CONTAINER COMPONENTS

Main Parts	Temperatures (degree C)		Cumulative Irradiation Dose (for 60 years)	
	Initial Storage	After 60 years	Neutron (n/cm^2)	Gamma (Gy)
Body & lids [*1]	182	84	2.12*10^{15}	1.04*10^{6}
Neutron Shielding Material (Resin)	163	79	4.13*10^{14}	6.34*10^{2}
(PG Water)	147	78	1.52*10^{14}	1.23*10^{3}
Basket	265	100	4.87*10^{15}	3.73*10^{6}
Metal Gasket	119	69	(Less than *1)	(Less than *1)

9.4. METAL GASKETS

Dry storage casks and canisters with bolted closures involve configurations that have metal or elastomer gaskets. The metal gaskets include:

- O rings: uncoated, stainless steel, Inconel, Nimonic steel.
- O rings: coated, including aluminium, silver, copper.
- Double O rings, including pressure monitoring in the space between the two seals.

The material of which gaskets are made is likely to affect their degradation in either normal service or in off-normal conditions. Residues from loading operations on the flange surfaces or condensate/moisture getting onto the outer gasket face during service are mechanisms which can initiate corrosion of the gasket.

Apart from normal transport/storage conditions metal gaskets have to with-stand the following ‘accidental’ conditions:

- Parallel sliding of the lid and the body, resulting in a lateral displacement of the contact surfaces
- Separation of the lid and the body, resulting in a decompression of the gasket
- 30 minutes at 800ºC fire on the cask, resulting in a peak temperature of the gasket.

9.4.1. France – evaluation of the thermal behaviour of spring-energized metallic gaskets

The use of spring-energized metallic gaskets in dual purpose casks is well known. They comprise (shown in Fig. 45.): A helicoïdal internal spring creating an internal sealing stress; an intermediary jacket which evenly distributes the spring force; an external coating in ductile material which compresses into the flange absorbing an inflections.

TN International has been evaluating new spring-energized metallic gaskets for application in their TN 85 and 24E casks. The gasket qualification programme comprised:

- Multiple compression / decompression tests
- Rotation tests
- Scrapping tests
- Very high temperature tests (accident conditions)
- Long term tests at high temperature

The results of these tests are provided [83]. The thermal aging tests are reported:

Fig. 45. Metal gasket for use in TN85 and 24E casks.

(1) Short-term high temperature tests

Thermal cycle has been evaluated on 2 gaskets in a test assembly (Fig. 46). The test cycle comprised raising the temperature from 160°C to 400°C min, holding the temperature for 1 hour above 300°C with a minimum peak temperature of 400°C, cooling to 160°C in 10 hours minimum.

The test temperature reached 407°C and pressure testing showed a leak rate of $< 10^{-10}$ Pa.m^3/s; which is considered as leak tight by the competent authority.

(2) Long-term temperature tests

Three seal test assemblies (3 x 2 gaskets) as shown in Fig. 46 were put into 3 ovens at the following temperatures: 150, 200, 250°C. The assemblies were removed at the following times 1 000, 2 000, 4 000 …10 000 hours and assessed for leak tightness.

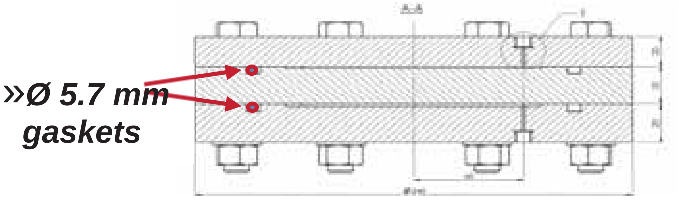


Fig. 46. Gasket test assembly.

Pressure testing for temperatures up to 250°C for greater then 10 000 hours has shown the leak rate to be less than 10^{-10} Pa.m^3/s; which is accepted as leak tight by a competent authority.

9.4.2. Japan – long term sealability tests

The confinement structure of the metal cask is designed to have a highly reliable multi-barrier system using metallic gaskets instead of the conventional rubber gaskets. Therefore, it is very important to clarify the influence of the stress relaxation of the gaskets on the containment performance of the metallic gaskets for a long-term usage. Long-term containment of the secondary metal gasket in full scale cask lid structures have been measured for more than 15 years under a constant temperature of 160˚C (Fig. 47.). The results indicate that containment will be maintained for more than 50 years, when decay heat of the spent nuclear fuel is taken into account.

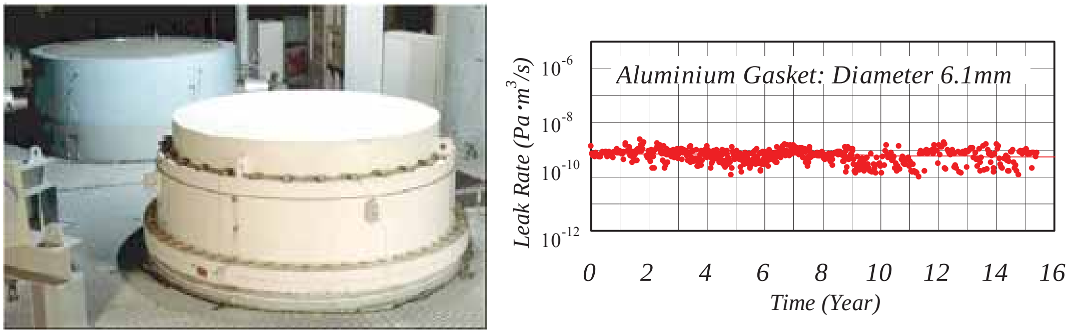


Fig. 47. Sealing tests with two full-scale lids.

9.5. POLYMERS USED TO SEAL LARGE CONSTRUCTIONS

In unlined water retaining structures water seapage at the expansion joints is prevented by the incorporation of water retaining flexible wide polymeric strips or ‘water bars’; for example polyvinyl chloride. The gaps between the joints are also filled with polymeric sealants for example poly sulphides or poly urethanes.

During their service life these polymers will degrade due to the combined effects of the radiation and the service environment (i.e. whether in air or in contact with the water) resulting in reduced mechanical properties and greater potential to fail. Accelerated testing has shown that polymers in contact with water were found to be serviceable after receiving an integrated gamma dose of $1x10^6$ Gy.

A secondary effect of polymer degradation is that species (polymer dependent) such as silicate, hydrochloride are released, where the polymers is in contact with the pool water these will be leached to the bulk pool water an d potential impact on water treatment systems.

The degree of degradation is effected by a number of parameters which include: Thickness; Compositon (presence of other materials); Dose rate; Medium they are in contact with (air, water, boric acid etc.).

It has also been shown that polymer degradation is influenced by dose rate; i.e. degradation will be greater if the material is irradiated at a low dose rate for a long time compared with the degradation produced by irradiation to the same total dose at high dose rate over a shorter period of time. Clough and Gillen [84] have developed a model which is able to predict the dose to produce a given reduction in properties of PVC cable insulation at low dose rates typical of service environments using radiation damage data obtained in accelerated tests at high dose rates.

(1) UK – water bar service life assessments

Thorp Receipt & Storage (Sellafield –UK) is currently being evaluated for the bulk storage of Advanced Gas Reactor fuel post reprocessing operations. Storage durations for up to 80 years are being evaluated. As the storage ponds in TR&S are only lined, with stainless steel, at the wind water line water retention at the expansion joints is reliant on polymeric sealants and water bars; which are life limited by radiation dose up-take.

The expansion joints comprise of three PVC water bars at different levels in the expansion joint step and a poly-sulphide sealant between the pond floor concrete screed and the first water bar.

Accelerated radiation testing of the PVC water bars used in the construction of TR&S showed that the material received a moderate to severe damage up to 1×10^6 Gy and was unusable after receiving a dose of 5×10^6 Gy; when irradiated in water at a temperature of 35ºC with an applied does rate of 10^4 Gy hr-1.

After considering a number of factors, for example low dose uptake correction, difference between irradiation in air and water etc. a safety margin of two orders of magnitude has been applied which leads to a working dose uptake limit of 2×10^4 Gy.

Based upon past and future fuel storage planning models, using gamma data derived from FISPIN calculations for the most demanding fuels that have and will be positioned over the expansion gap, the following have been established:

- The dose uptake to date and hence an indication of the current condition
- An estimate of the lifetime dose uptake by the water bars for the proposed new duty
- If the lifetime integrity is likely to be compromised to suggest shielding requirements

While it has been demonstrated that a number of the water bars in the expansion gaps will not approach the 2×10^4 Gy limit, there is potential for it to be exceeded in one area. A number of measures can be introduced to manage this position; for example the area is only used for low irradiation long cooled fuel. A number of shielding option shave also been suggested these include storing empty storage containers over the expansion gap or in the inclusion of 30mm thick steel plates.

9.6. STAINLESS STEEL

Stainless steel components are highly resistant to corrosive attack due to the formation of protective oxide films. For example, corrosion as a consequence of pitting or galvanic attack has been shown to be negligible [85], and the rates of general (or uniform) corrosion only become significant over extended time periods (100s of years). To put the later in context the measured corrosion rate of stainless steel in radiolysed water is 0.3 μm/year and <0.1 μm/year in a caustic environment.

In terms of radiation hardening the cumulative neutron dose for steels in storage application of around 10^5 n/cm^2/s is too low to be of concern; embrittlement occurs for cumulative neutron doses >10^{17} n/cm^2/s).

The most significant failure mechanisms affecting Stainless Steel, under the right conditions, are stress corrosion cracking and microbial failure. Pitting, galvanic or crevice corrosion are considered to be negligible.

(1) Stress corrosion cracking

For a sensitized stainless steel, when the chromium content in the grain boundary has fallen below 12%, subjected to stress is brought into contact with aggressive ions, such as chloride, then stress corrosion cracking will result. Sensitization of stainless steel can result by one of two mechanisms:

(2) Thermal sensitization

In a certain temperature range, the chromium in unsterilized stainless steels can migrate from grain boundaries and combine with any free carbon in the metal to form particles of chromium carbides. Thermal annealing processes are sufficient to re-establish a homogeneous distribution of chromium and thus remove thermal sensitization.

(3) Radiation induced sensitization

Changes in the concentrations of chromium at metal grain boundaries can occur due to high-energy (fast) neutrons (and does not involve the formation of chrome rich carbides). Because of the irradiation profile along a fuel stringer or fuel assembly, the effect peaks in certain zones. Unlike thermal sensitization, the sensitized microstructure remains as an artefact of reactor operation after the fuel is removed from the reactor.

(4) Microbial attack

Over time, microbial colonies may become attached to pipes etc., establishing local environments that favour the growth of species, which would not otherwise survive. Established colonies are also highly resistant to fluid forces and most chemical treatments, making removal difficult. The microbial colonies effectively shield the metal surface from water treatment chemicals, rendering corrosion inhibitors ineffective.

It has been demonstrated that welding of austenitic stainless steel increases their susceptibility to microbal induced corrosion (MIC) compared to smooth pipe surfaces. In fact, the weld and the associated heat-affected zones become the primary area of MIC attack.

The impact of MIC has generally been ignored as it is hard to detect and this mechanism is usually masked by general corrosion. The only indicator of its influence is where components under periodic replacement programmes have to be replaced at slightly greater frequency than originally anticipated.

9.6.1. Japan – metal canister stress corrosion cracking tests

The siting of spent fuel interim storage facilities in Japan are likely to be at coastal locations. Temperature decreases during storage period and salt condensation increases on the metal canister surface. Key issue for realization of the metal canister storage technology should be ‘Long-term integrity of canisters’, considering deterioration of metal canister in a salt water environment. As an austenitic stainless steel may be vulnerable for SCC under a certain unique set of circumstances, it is important to prevent penetration through the wall thickness by SCC. Factors affecting SCC corrosion in salt water environment are shown in Fig.48. Therefore, the device which can reduce a salt particle into storage buildings has been developed and CRIEPI has proposed a salt particle collection device with a low flow resistance which does not interfere with the air flow into the storage facility. The effect of the device was evaluated quantitatively in basic experiments and in the field tests. Data to demonstrate integrity of canister have been continuously obtained. Candidate canister materials did not fail for 54 300h by constant load test, which simulated residual stress of weld [86].

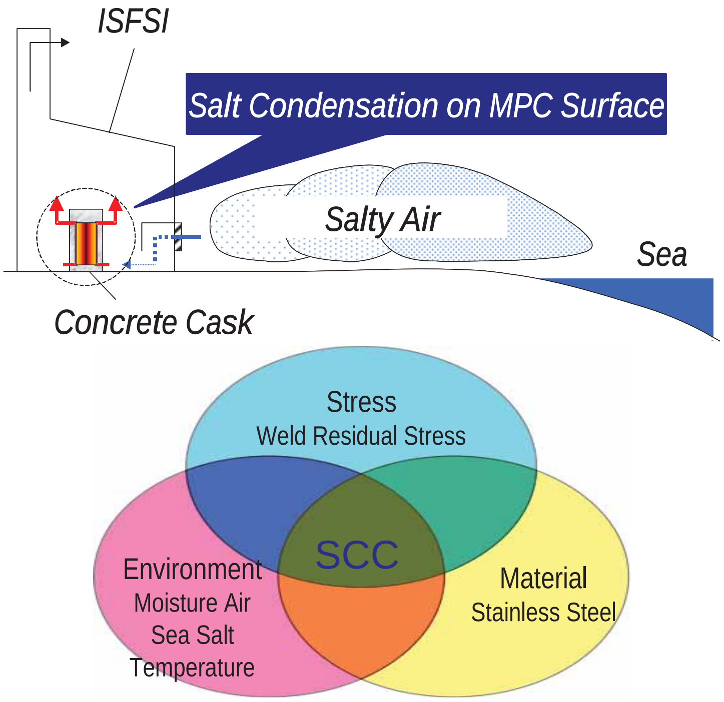


Fig. 48. Deterioration of metal canister in a salt-water environment.

9.6.2. Slovak Republic – evaluation of stainless steel used in wet storage

Spent fuel from the nuclear power plant Jaslovske Bohunice is kept in wet storage. The spent fuel assemblies are placed in racks and stored in large water pools. This type of storage requires a long term corrosion stability of the structure materials used. Monitoring methods have been developed in order to be able to maintain integrity and functionality of the storage components.

The main structural material in the spent fuel storage is Cr-Ni austenitic stainless steel stabilized by Ti, grade 08Ch18N10T, equivalent to AISI 321. This material is used in pools lining, piping, pumps and valves. Austenitic stainless steels stabilized by Ti have very good plastic properties at low temperature and high values of notch impact strength. These materials have very good welding characteristics as well as resistance to inter-granular corrosion attack. The uniform corrosion of this steel and its weld joints in water with high purity is negligible. Nevertheless, the potential of other forms of localized corrosion, for example stress corrosion cracking (SCC), pitting and crevice corrosion, need to be considered.

In the upgraded storage facility, a new grade of stainless steel is used. The steel is called 'ATABOR' and is in based on the AISI 304 stainless steel in an alloy with boron. The corrosion resistance of the ATABOR is similar to this of steel AISI 304. The chemical properties of the mentioned steels are shown in Table 8.

Cooling water in the spent fuel pools is controlled by measuring **specified** control parameters; typical values the operating limits are provided in Table 9.

TABLE 8. CHEMICAL COMPOSITION OF STRUCTURAL MATERIALS

STEEL	Chemical Composition (mass %)								
	C	Mn	Si	Cr	Ni	Ti	P	S	Boron
08Ch18N10T	Max. 0.08	Max. 2.0	Max. 1.0	17.0 – 19.0	9.5 – 12.0	Min. 5.0	Max. 0.045	Max. 0.030	
ATABOR	Max. 0.03	Max. 2.0	Max. 0.8	18.0 – 20.0	10.0 – 12.5	------	Max. 0.035	Max. 0.025	Min. 1.1

The corrosion monitoring specimens as shown in Fig. 49 were prepared from all three materials and placed in the spent fuel pool cooling water. Besides stainless steel plates with weld joints of 08Ch18N10T and ATABOR, characteristic old parts of the equipment were placed into the pool.

TABLE 9. MEASURED PARAMETERS AND TYPICAL VALUES IN THE COOLING WATER

Parameter	Units	Limiting Value	Typical Values
pH	-	5.5 – 8.0	5.8 – 6.5
Cl^-	mg/kg	<0.1	<0.02
Conductivity	µS/cm	<3	1 – 1.5
Suspended Particles	mg/kg	<0.5	<0.4
^3H	Bq/m^3	<3.7*10^9	10^5
Total Activity	Bq/m^3	<4.1*10^7	10^2 – 10^3
Temperature	^{0}C	<50	20 – 50

Table 10 shows the results obtained from corrosion monitoring of specimens.

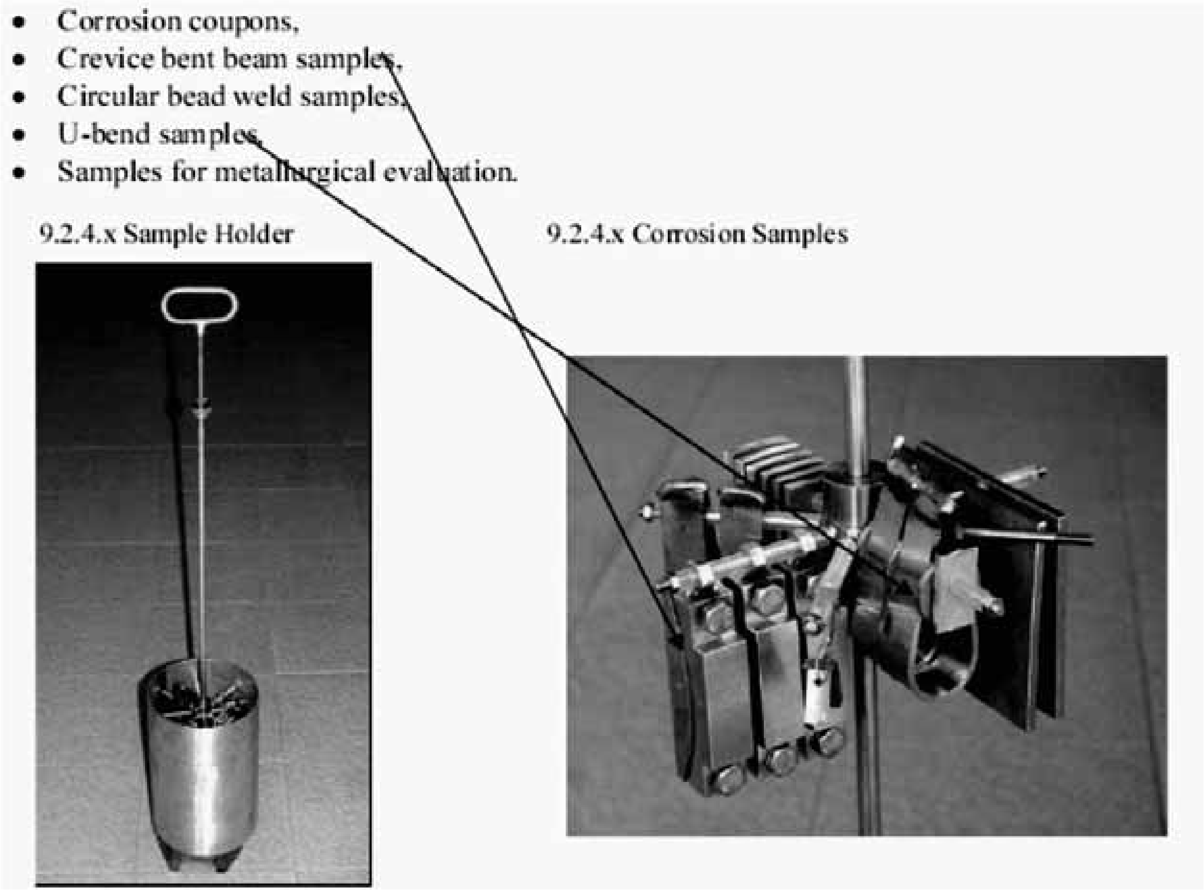


Fig. 49. Corrosion specimens.

TABLE 10. EXAMPLES OF THE RESULTS OBTAINED BY CORROSION MONITORING OF SPECIMENS

		Weight			
Sample No.	Position	Before exp. [g]	After exp. [g]	Difference [g]	Weight Loss [g.m^{-2}/year]
1-1	Surface	96.7234	96.7195	0.0039	0.1094
1-2	Surface	96.4301	96.4265	0.0036	0.1010
1-3	Surface	96.1962	96.1924	0.0038	0.1086
1-4	Beneath Surface	96.8849	96.8817	0.0032	0.0898
1-5	Beneath Surface	98.0284	98.0284	0.0045	0.1263
1-6	Beneath Surface	96.4244	96.4190	0.0054	0.1516

The main conclusion from samples removed for examination after 1½, 3, 4, 5 and 6 years was the maximum weight loss was < 0.8 g.m^{-2}.y^{-1} and there were no signs of corrosive attack.

9.7. CARBON STEEL, BEHAVIOUR IN DRY STORAGE

Carbon steel is an iron based alloy with 0.05–1% carbon and small amounts of other elements, principally manganese (~1%). While it did find widespread use coated in wet fuel storage rack applications it has since been replaced by stainless steel or aluminum.

Carbon steel is primarily used in large forgings for transportation casks and dry storage technology, including storage cask bodies, components and over packs.

Degradation of exterior carbon steel surfaces exposed to the atmosphere is minimized with coatings that are subject to periodic inspection and repair as required. Carbon steel surfaces inside the cask seals are often protected with a metal coating, generally zinc or aluminum. Otherwise, deterioration of the steel surfaces can be minimized by effective removal of reactive species, for example water and oxygen, in the drying process. The integrity of the cask seals and/or welds is essential to preclude ingress of moisture or air.

9.7.1. Japan – metal storage cask tests

(1) Drop test without impact limiters [87]

In an interim storage facility of spent fuel, metal casks will be handled without impact limiters. Although there have been a lot of tests and analyses reported for evaluation of drop tests of metal casks, no quantitative measurement has ever been made for any instantaneous leakage through metal gaskets subjected to the impulsive loads. In this study, leak tests were performed using a full-scale metal cask without impact limiters considering drop accidents during handling in a storage facility. Figure 50 shows a drop test with the full-scale metal cask. A series of drop impact tests (a horizontal drop test from a 1m height and a rotational impact test around an axis of a lower trunnion of the cask from the horizontal orientation at a 1m height) were conducted onto the reinforced concrete slab simulating the floor structure of the facility. The main measurement items are the sliding and opening displacements of the primary and secondary lids, leak rates, and inner pressure in the volume between two lids. Instantaneous leak rate was quantitatively measured at the drop tests.

Fig. 50. Drop test with the full-scale metal cask.

(2) Airplane crash test

To evaluate damage of cask in airplane crash event at a storage facility, simulated crash tests are being planned. Simulated engine of airplane will hit a model cask as shown in Fig. 51. Leak rate from the metallic gasket in the cask will be measured at the impact in these tests.

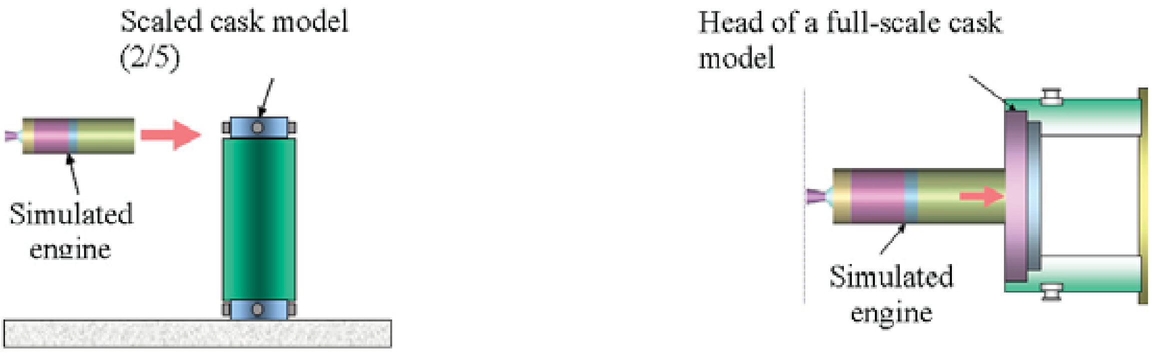


Fig. 51. Simulated crash tests.

9.7.2. UK – continued operations - machines utilization

Thorp Receipt and Storage (at Sellafield, UK) is currently being investigated as a bridging solution for the storage of AGR fuel pending the out-come of a national review into spent fuel management. To enable continued operations, a mechanical assessment of the major machines utilization has been carried out in order to establish if these machines will require replacement from an utilization point of view in the next 80 years.

The past and projected utilization of all the TR&S handling machines has been assessed with no major issues identified. The most utilized pieces of equipment have been identified as the 125t flask lifting beams that could potentially use 64.5% of the design code allowable fatigue cycles over the next 22/23 years in order to complete the storage phase of the project.

10. MONITORING TECHNIQUES

In IAEA-TECDOC-1343 [4] the purpose of performance monitoring was outlined and brief descriptions of monitoring technologies in routine use were given. Additional Member State experience and practice in monitoring and surveillance is also reported in IAEA-TECDOC-461 [88].

The current phase of the SPAR coordinated research programme reports monitoring activities in participating countries. The main aim of these technologies are to confirm on-going spent fuel integrity or to provide an early indication of developing conditions which would impact on the long–term spent fuel integrity if no corrective action is taken.

10.1. METHODS FOR CONFIRMING PERFORMANCE

The techniques employed range from basic visual inspections (through the biological shielding wet or dry (cave)) to sophisticated radiometric systems (such as a burn-up monitor).

Examples of commonly used techniques in participating countries are summarized in Tables 11 and 12.

TABLE 11. SUMMARY OF MONITORING TECHNIQUES AND APPLICATIONS

Technique \ Function	Containment	Shielding	Criticality	Operability	R&D Data Confirmation	Other Safety Functions
Pressure Monitoring	√				√	
Fission Gas Sampling	√					
Temperature				√	√	√
Radiation Detection Systems		√				√
Liquid Sampling	√			√		√
Visual Inspections			√	√		√
Gas Sampling (non-active)					√	
In-pool Inspection Systems					√	√
Burnup			√		√	√
Ultrasonic Test	√			√		

TABLE 12. SUMMARY OF CURRENT AND FUTURE MONITORING TECHNIQUES AND APPLICATIONS IN EACH COUNTRY

TECHNIQUE/FUNCTION	**Argentina**		**Canada**		**France**		**Germany**		**Hungary**		**Japan**		**Slovak**		**Spain**		**Sweden**		**U.K.**		**U.S.A**	
	W	**D**	**W**	**D**	**W**	**D**	**W**	**D**	**W**	**D**	**W**	**D**	**W**	**D**	**W**	**D**	**W**	**D**	**W**	**D**	**W**	**D**
Pressure Monitoring	---	F	---	C	---	C	---	C	---	C	--	C	--	F	--	C	---	---	--	--	--	C
Fission Gas Sampling	---	F	---	---	C	--	C	C	---	C	--	C	C	F	C	--	C	---	--	C	C	--
Temperature	C	C	C	C	C	--	C	C	C	C	C	C	C	--	C	C	C	---	C	C	C	--
Radiation Detection Systems	C	C	C	C	C	C	C	C	C	C	C	C	C	F	C	C	F	---	C	C	C	C
Liquid Sampling	C	---	C	---	C	--	C	---	C	---	C	--	C	--	C	--	C	---	C	--	C	--
Visual Inspections	C	---	C	C	C	--	C	---	C	---	C	C	C	F	C	C	C	---	C	C	C	C
Gas Sampling (non active)	---	F	--	C	--	--	---	C	---	C	--	--	--	F	--	--	C	---	C	--	--	C
In-pool Inspection Systems	C	---	C	---	C	--	C	---	C	---	C	--	--	--	C	--	C	---	C	--	C	--
Burn-up	---	---	C	F	--	--	---	---	P	F	F	F	F	--	--	F	F	---	P	--	--	F
Ultrasonic Test	---	---	C	--	--	--	---	---	C	--	C	--	--	--	C	--	--	---	--	--	C	--
Corrosion probes	---	---	--	--	--	--	---	---	---	--	--	--	--	--	--	--	---	---	F	--	--	--

Note: W (Wet), D (Dry), C: Current, F: Future, P: Prior to reprocessing only, Most techniques for confirming spent fuel integrity during storage are retrospective or reactive; i.e. when you detect radionuclides for example caesium in pool water or krypton in the cover gas the spent fuel has already failed. The plant operator then has to establish where the failure is, under take some form of failure analysis and then make a decision on how to manage the failed fuel.

10.1.1. Technologies for confirming on-going spent fuel integrity in wet storage

The confirmation of on-going spent fuel integrity in wet storage is routinely monitored by liquor sampling for the mobile fission products ^{134}Cs and ^{137}Cs. The ratio of ^{134}Cs to ^{137}Cs provides an indication of whether a fuel failure has occurred or where there is multiple fuel failures already present whether new fuel failures are occurring. The problem with this approach is that once cesium is detected it is usually too late. For some fuels/components, for example stainless steel, where storage is in dematerialized water an early indication of deteriorating conditions can be monitored through sampling for elemental nickel and for the activation products ^{54}Mn, ^{55}Fe, ^{60}Co and ^{63}Ni. If LWR fuel is present detection of evolving storage conditions is hampered by interference from CRUD.

The use of on-line corrosion probes fabricated from the fuel clad or probes designed to simulate known fuel corrosion mechanisms provide a mechanism for bridging the gap between detecting failure and conditions that would lead to failure; in turn this gives the operator an opportunity to take corrective action. Techniques which have been investigated include: Electrochemical Noise, Electrochemical Impedance Spectroscopy, Zero Resistance Ammeter, Field Signature Method and Ratio-metric electrical resistance.

The use of on-line corrosion probes is of particular interest for spent fuels which are known to be susceptible to failure under abnormal operating conditions; although Zircaloy-4 probes have been used to study the impact of changing pool chemistry on LWR fuel. Further development and maturity of these technologies, may offer application in providing supporting data to re-licensing or safety case review activities; as an alternative to under taking post storage examination of long stored fuel or fuel storage systems.

A specific example of the application of electrochemical noise (EN) and electrochemical impedance spectroscopy (EIS) for confirming Magnox fuel passivity is presented later.

10.1.2. Technologies for confirming spent fuel integrity in dry storage

Apart from the MVDS technology where the cover gas of individual storage tubes can be sampled in-situ, by portable gas analysers, for the presence of fission gas such as krypton or air ingress oxygen, confirmation of continued spent fuel integrity during storage for all other dry storage technologies would require the containment to be broken and the fuel to be sentenced for hot laboratory inspection.

On-going spent fuel integrity for these systems is inferred through external temperature and containment monitoring (i.e. pressure monitoring of the seals and radiological/visual inspections of the outer cask fabric), quality assurance during system closure and supporting R&D that provides the confidence that failure during storage can be discounted.

To provide further reassurance to key stakeholders that the spent fuel is still intact during storage a number of non-intrusive techniques (gamma scanning and ultrasonic) have been proposed. Both of these proposals rely upon a modified lid arrangement to facilitate monitoring through the shielding.

- ***Gamma Monitoring***

Probably one of the most attractive options for monitoring the condition of spent fuel in dry storage is by direct measurement of the 514keV ^{85}Kr γ line. This technique is particularly attractive proposition for monitoring the condition of spent fuel during the early years of dry storage (which are probably the most onerous for Zircaloy clad fuels). If storage duration were to extend to 50+ years then the technique becomes less attractive given the half-life for ^{85}Kr (10 years).

The feasibility of this technique is described in more detail in Chapter 10.2.3.

- ***Acoustic Monitoring***

Monitoring the velocity of sound through the cover gas is an alternative mechanism for detecting the condition of the atmosphere inside a sealed system. Such a system was proposed for the TranStor^{TM} Storage system [89] for the detection of oxygen in helium (indicator of air ingress) but, was not developed into a fully operational system.

10.2. COUNTRY SPECIFIC MONITORING EXPERIENCES

10.2.1. Temperature monitoring of dry storage casks in Canada

To confirm that the dry storage casks (DSCs) would not suffer from freeze-thaw damage to a significant extent, a programme to monitor the temperatures of both the storage facility and the DSCs was put in place. The monitoring took place at the Used Fuel Storage Facility – Phase 1 of the Pickering Station from the latter part of December, 2000, to the end of March, 2001, coinciding with the return of above freezing temperatures. The purpose of the monitoring was to record the minimum temperatures that could be experienced by the DSCs. To fulfill this objective, a total of 31 thermocouples were placed in various locations: outside of the storage facility to measure ambient air temperatures, indoors within the storage facility including the concrete floor surface, on the surface of the DSC's outer shell for DSCs most exposed to the coldest temperatures, and, thermocouples located at the bottom of the DSCs to measure their temperature in contact with the concrete floor.

The observed temperatures indicated that the temperature outside of the storage facility on the coldest day was -17.5 ºC, and, there were 31 freeze-thaw cycles over the monitoring period. Yet, at all times, concrete floor temperatures under the DSCs remained above zero, as well as the DSC's outer shell temperatures being about 2 ºC above zero. In the facility itself, the lowest air temperature reached -4.5 ºC and the lowest exposed concrete floor temperature was -0.5 ºC. Inside the storage facility, ambient air between two DSCs freeze-thaw cycled seven times, while the most exposed wall of the facility freeze-thaw cycled four times.

The results of that monitoring showed that the storage facility provides significant protection against freeze-thaw conditions. Data indicated that the DSCs remained above zero at all times even though ambient air temperatures within the facility were on seven occasions below freezing. However, the low temperatures recorded inside the facility would suggest that as the stored fuel decays, the DSCs might experience some freeze-thaw cycles during their 50 year design life. In view of the 6% air-entrainment provided in the concrete, some freeze-thaw cycling of the concrete is not viewed to leading to a detrimental impact.

10.2.2. Development of an inspection technique for the confirmation of spent fuel integrity during storage in Japan

In Japan a non-intrusive method for determining spent fuel integrity in dry storage canisters has been investigated [90]. If a spent fuel rod failed during storage in a canister the gaseous fission products (FP), including ^{85}Kr, would diffuse into the canister cavity. Through gamma scanning, from outside of the container (Fig. 52), for the distinctive 514 keV gamma ray which ^{85}Kr emits it is possible to detect if failed fuel is present.

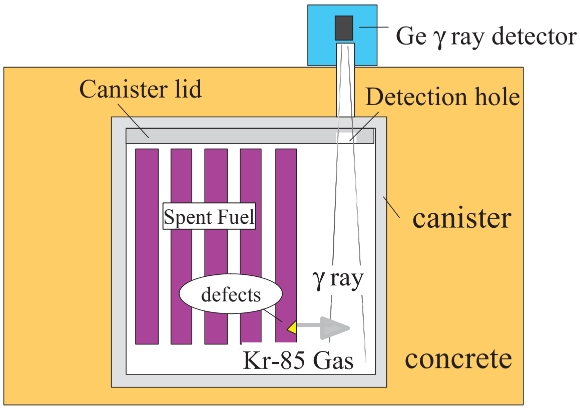


Fig. 52. Concept of non-destructive monitoring technique.

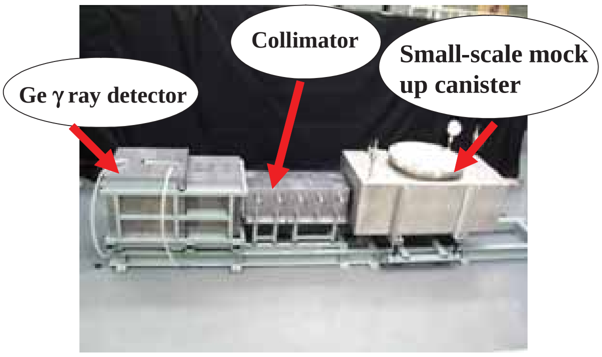


Fig. 53. The whole view of the mock-up equipment using ^{85}Kr gas.

The experiments were performed using a small-scale mock-up canister and ^{85}Kr gas (Fig. 53) to establish the capability to detecting ^{85}Kr gamma emission in the high radiation back-ground of the spent fuel (noise). Figure 54 shows an example of a gamma scan using a high resolution Ge detector. It shows that the gamma ray of ^{85}Kr (514 keV) can be distinguished from the gamma ray of positron annihilation (511 keV). Data fitting enhances the extraction of the '^{85}Kr signal' from the high radiation background of the spent fuel.

The Measurement limit function of ^{85}Kr was acquired from this experimental data (Fig. 56). This shows the lower limit of detection for ^{85}Kr from the peak ratio of the gamma ray (2) of ^{85}Kr to the gamma ray (1) of positron annihilation as a function of the base ratio of it to gamma ray of Compton scattering (3) in Fig. 55.

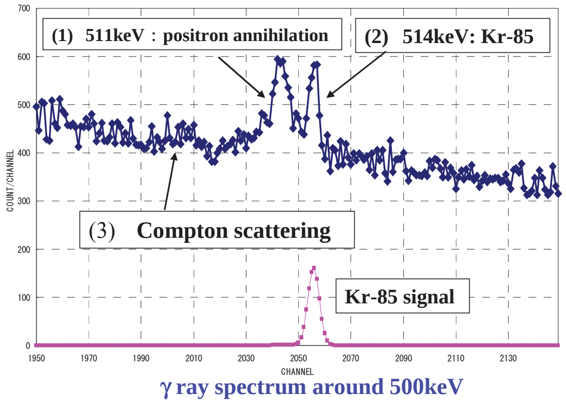


Fig. 54. Example of gamma ray spectrum of the mock-up.

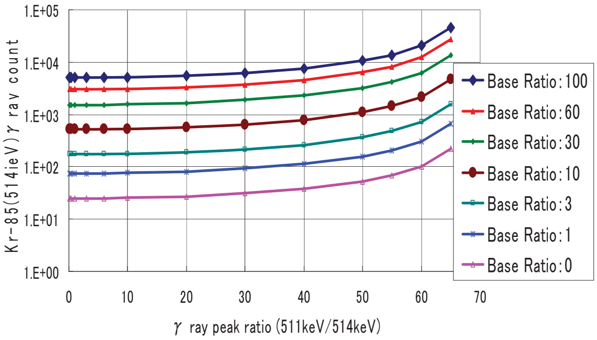


Fig. 55. Measurement limit function of ^{85}Kr examination.

The applicability of the non-destructive monitoring method has been evaluated for a full scale canister loaded with 21 PWR fuel assemblies. This showed that shielding by the canister lid impacted on gamma detection efficiency outside the canister. To improve the detection efficiency a new lid design with a penetration to half the wall thickness (10 mm) was incorporated (Fig. 56).

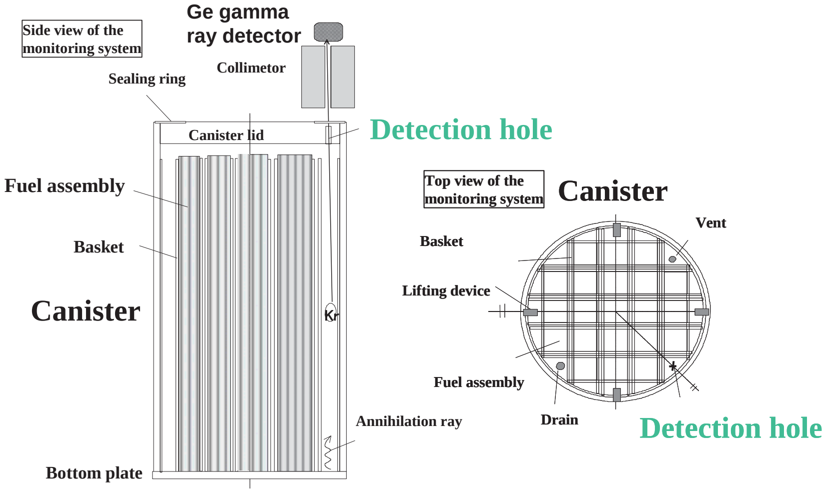


Fig. 56. Equipment of detection hole in the lid of storage canister for ^{85}Kr γray detection.

The detection performance of this set-up has been analysed and it was found that a failed fuel rod could be detected above the background gamma rays (as out-lined above) if a minimum of 10% of the fuel rod ^{85}Kr content is released (Table 13).

TABLE 13. THE DETECTION PERFORMANCE OF GAMMA RAYS

^{85}Kr release	7.0E+09 Bq	7.0E+10 Bq	7.0E+11 Bq
% of ^{85}Kr in a fuel rod	1%	10%	100%
514 keV peak (cps)	2.98E-03	2.98E-02	2.98E-01
Measuring time			
3 hours	nd*	detection	detection
28 hours	nd	detection	detection
280 hours	nd	detection	detection

*nd: cannot detect

The application of this monitoring technique has been considered as an inspection method prior to the transportation of spent fuel from the interim storage facility to the reprocessing plant. Given that it is necessary to repack a canister into a transport cask at the time of transportation, establishing that the fuel is still intact using gamma scanning has been proposed for the canister repack area of the storage facility (Fig. 57). A gamma scan of around 24 hours/canister has been derived; prior to canister transportation to the reprocessing plant.

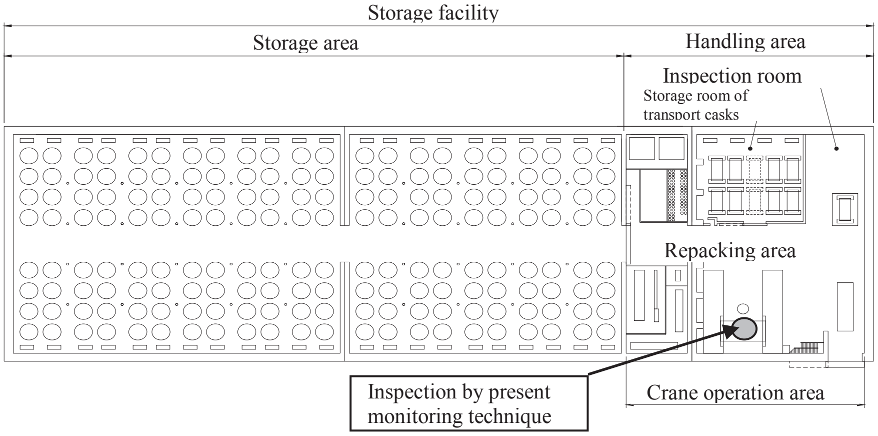


Fig. 57. Plan of interim storage facility showing concrete cask inspection bay.

10.2.3. Monitoring of pool lining integrity by Acoustic Emission in the Slovak Republic

In the Spent Fuel Interim Storage (SFIS) facility at the nuclear power plant Jaslovské Bohunice the integrity of the pool lining is monitored periodically by the use of acoustic emission.

The technique involves listening to the natural energy waves released when a material undergoes plastic deformation. In the case of a metal pool liner this results from changes in load, pressure or temperature. It is a qualitative technique for establishing damage within a structure and is used for monitoring the development of active defects. Defects are detected as the stress levels (and corresponding energy levels released) in front of the crack tip are several times higher than the surrounding metal.

At the SFIS there are six sensors installed in each pool and measurements are performed periodically using a portable data collection system. Comparison with archived data is used to establish if there any cracks or defects developing. To date there has been no significant defects detected.

10.2.4. Monitoring of Magnox fuel passivity during modifications to the Oldbury NPP pool chemical clean-up system in the United Kingdom

A Magnox corrosion probe fabricated from A180 clad has been deployed over a 65 week period at Oldbury NPP (see Fig. 58) to monitor Magnox fuel clad passivity during modifications to the pool chemistry system. The system was developed by National Nuclear Laboratory (UK).

Fig. 58. A Magnox corrosion probe system.

Two types of electrochemical techniques were employed:

- Electrochemical impedance spectroscopy (EIS)

EIS works by applying a low amplitude sinusoidal potential excitation to the working electrode via the reference electrode and measuring the current response against the counter electrode. From the frequency response data Nyquist plots for the system can be generated. Typical Nyquist plots of EIS data for Magnox Al80 cladding for different storage conditions (Sodium Hydroxide dosed demonized between pH 11 and 12) are shown in Fig. 59.

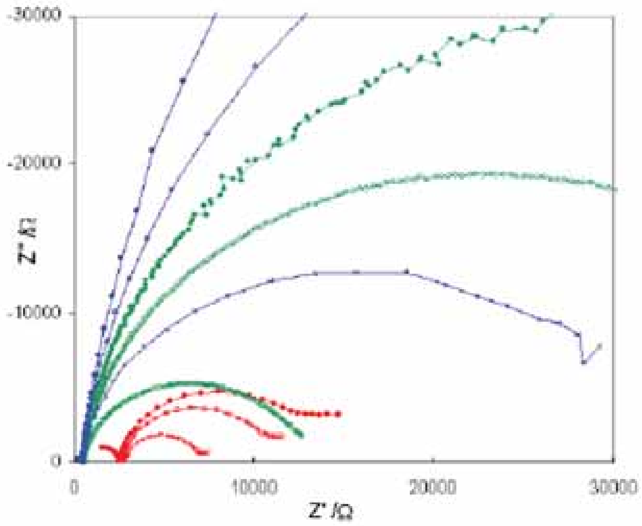


Fig. 59. Typical Nyquist plots of electrochemical impedance spectroscopy.

- Electrochemical Noise (EN)

EN is particularly sensitive for identifying the early stages of localized corrosion, it measures the small fluctuations in the potential and current caused by microscopic variations in the corrosion processes occurring on a freely corroding electrode. Examples data generated by EN for Magnox A180 cladding under passive and pitting attack are shown in Figs 60 and 61.

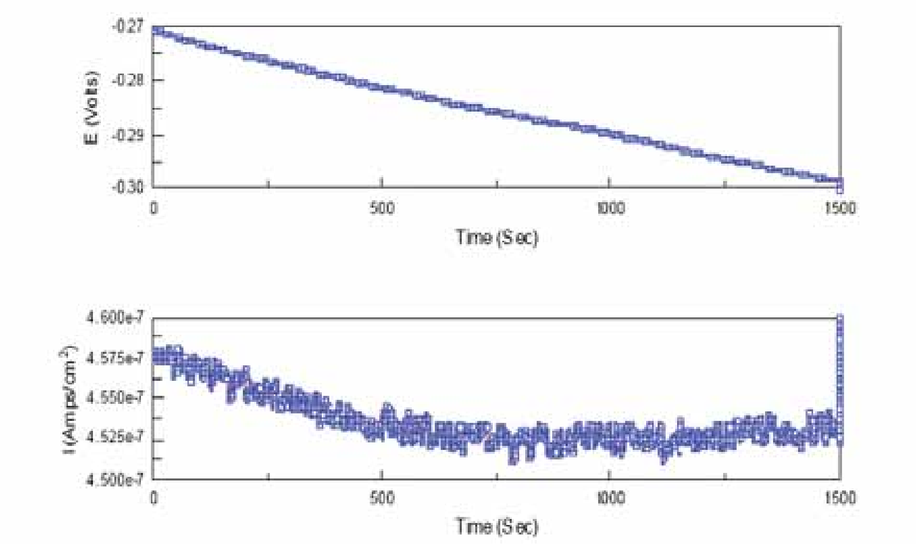


Fig. 60. 1st Example of Electrochemical Noise (EN) data for Magnox A180

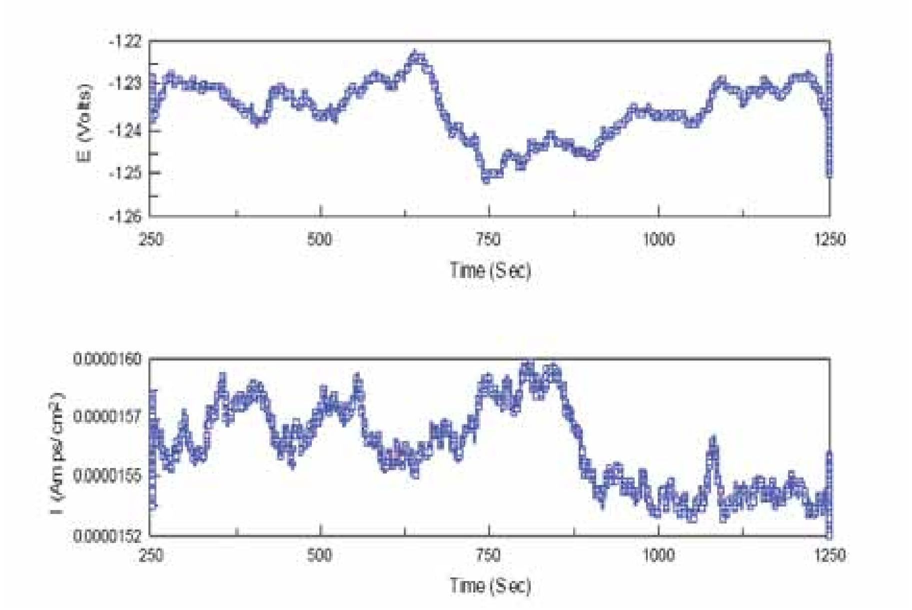


Fig. 61 2nd Example of Electrochemical Noise (EN) data for Magnox A180

During the 65 week installation the corrosion probe was configured between EIS and EN every few weeks. A comparison of the data collected with the laboratory data (shown above) showed that the EIS Nyquist plot data remained in the high pH 11.7 to 12 states and the EN data remained in the passive state; i.e. low galvanic current and low noise with potential. The results confirmed that the Magnox fuel passivity remained unaffected during the modifications to the pool chemistry; as had been expected from the pre-job risk assessment.

10.2.5. Monitoring of dry storage cask integrity in the United States

Standard monitoring techniques are used in conjunction with the use of All-metal and Concrete/Canister storage systems. Dry storage cask inspection monitoring activities typically consist of:

- Pressure Monitoring (Bolted Closure Systems Only)

The pressure of the cover gas is monitored (at various locations, depending on the cask type) to verify the integrity of the seals in the dry storage cask closure covers. Seal degradation resulting in loss of integrity would be detected by changes in monitored pressure.

Six instances of seal replacement since the placement of the first dry storage cask were documented at the Surry site. One occurred on a CASTOR X/33 dry storage cask secondary lid. The other five occurred on lid seals for the TN-32 dry storage cask. An evaluation of the TN-32 lid seal failures identified a design problem involving moisture intrusion under the environmental cover. This problem was corrected with a new weather cover design and different seal material.

- Visual Inspections (All-metal Casks)

The condition of the exterior of each dry storage system is inspected visually to ensure that the intended function of the casks exterior subcomponents is not compromised. Visual inspections look for signs of deterioration (corrosion) of the dry storage cask surface, and the inspections identify any debris accumulating on the dry storage cask surfaces. Debris can create the potential for localized conditions that support and enhance the corrosion process. Effects that are monitored by these inspections are loss of material, i.e., general corrosion and pitting. Additionally, normally inaccessible areas of the casks can be visually inspected on an opportunistic basis, for example, if they are lifted in preparation for movement or if an environmental cover is removed for maintenance. Since the normally inaccessible area made of the same materials as the exterior of the cask, inspections of the accessible areas provide an indication of the conditions of inaccessible areas.

Inspections of the dry storage casks have identified minor cases of corrosion or coating degradation (a precursor to corrosion). At the Surry site, approximately 40 dry storage casks underwent six visual inspections each during 2000-2001. As a result, a total of five work activities were initiated to repair coatings on four dry storage casks.

- Surveillance of Air Inlets and Outlets (Storage in Welded Canisters with Concrete Overpack Systems)

The air inlets and outlets of the concrete over-pack are visually inspected on a frequency based on analyses of normal and accident conditions to ensure that component temperatures remain below limiting conditions of the license.

Periodic observations of partial blockages due to small animals attracted by the heat emanating from the storage systems have been observed.

- Radiation Monitoring

ISFSI Technical Specifications typically require quarterly radiation monitoring at the ISFSI perimeter fence. Health Physics procedures implement this requirement through continuous monitoring at the ISFSI perimeter fence using Thermal Luminescent Dosimeters (TLD), for example, as well as quarterly radiation surveys at the Independent Spent Fuel Storage Installation (SFSI) security and perimeter fence locations.

- Note on License Extension

Conditions for license extension of the Surry and H.B. Robinson ISFSIs from 20 to 60 years have not resulted in additional monitoring except for the (1) polymeric neutron shield materials for which radiation monitoring activities were added; and (2) cask bottom for which an additional visual inspection (to be performed again in ~20 years) of the oldest CASTOR V/21 cask bottom was added to assess the condition of the breech plate bolts and cask bottom [91].

CONCLUSIONS

- Presently, spent fuel is mostly stored wet in at-reactor (AR) or away-from-reactor (AFR) facilities. Because many AR pools are approaching their full capacity, even after extensive re-racking, storage of spent fuel under dry and inert atmosphere is being used increasingly.
- Member States continue to deploy increased storage capacities successfully, with both wet and dry approaches considered to be fully mature technologies. Wet and dry storage continues to demonstrate good performance and provide system flexibility pending further progress in the back end of the nuclear fuel cycle.
- The storage technology (particularly dry storage) is undergoing a rapid evolution: new fuel and material design changes are coming on stream and target burnup were steadily increasing. These changes require continued analyses and potential adaptations of the storage technologies currently used, especially taking into account that spent fuel may have to be stored for extremely long periods of time; e.g. for up to 100 years and beyond.
- Wet storage continues to dominate as the primary method for storing spent nuclear fuel; >80% of all spent fuel is wet stored. The benefits provided by this technology are mainly associated with cooling efficiency and shielding. Also, it facilitates safeguards and one-off fuel inspection/examination exercises. Operating experience gained since the 1950s shows spent fuel storage performance for all types of fuel has been excellent.
- Almost 25 years of favorable experience exists with the dry storage of spent power reactor fuel and the quantities being placed into dry storage are increasing significantly. Since its conception, dry storage of spent fuel has evolved into a wide variety of systems including concrete canisters, steel-lined concrete containers or casks, and concrete vault-like structures. The results of the research indicate that fuel can be stored safely under the present conditions for many decades.
- In-reactor fuel integrity is determined by the measurement of activity coming from leaking fuel and the criteria for damaged fuel is primarily based on online monitoring and sipping systems which detect rod cladding failures. The onset of a fuel failure is usually detected by monitoring the gamma-activity level associated with specific radioactive fission products in the reactor coolant or off-gas. Dry storage cask inspection monitoring activities typically consist of pressure monitoring, temperature monitoring, visual inspection, surveillance of air inlets and outlets, and radiation monitoring.
- The potential degradation mechanisms that may affect cladding integrity during wet storage are uniform corrosion, pitting, galvanic, and microbiologically-influenced corrosion and hydriding. Potential degradation mechanisms that may affect cladding integrity during dry storage and subsequent handling and transportation operations are air oxidation, thermal creep, stress corrosion cracking (SCC), delayed hydrogen cracking (DHC), hydride re-orientation, hydrogen migration and re-distribution.
- Among the potential degradation mechanisms, in particular, hydride re-orientation has the potential to impair the ability of the cladding to effectively withstand potentially adverse mechanical challenges resulting from handling or transportation accidents. Much experimental work has been performed to get a better understanding of the conditions under which re-orientation can occur. Motivation for the work is for obtaining a better understanding of the potential effects of dry storage on cladding integrity during subsequent handling and transportation operations.

- The drying process is of paramount importance to the dry storage systems as it prevents the presence of any oxidizing gases and hydrogen from radiolysis. Temperature limits during drying have to be kept sufficiently low to minimize the potential for hydride re-orientation. If handling procedures designed in accordance with the fuel assembly mechanical design are strictly followed, no mechanisms exist that might cause the failure of fuel elements during a transition from wet-to-dry.
- All external surfaces and interfaces of storage facility components are susceptible to environmental attack. There have been active research programmes on the behavior of reinforced concrete, neutron absorbers used in storage racks and baskets for criticality control, neutron shielding materials, polymer seals, metal gaskets, stainless steels and carbon steels in wet and dry storage.
- It remains important to confirm the continuing viability of spent fuel storage by continuing to study and share results regarding fuel and materials behaviour in storage through efforts such as the Spent Fuel Performance Assessment and Research (SPAR-II) CRP.

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GLOSSARY AND ABBREVIATIONS

AFR	Away from reactor storage
AR	At reactor storage
AGR	Advanced Gas Cooled Reactor
BEFAST	**Be**haviour of Spent **F**uel **A**ssemblies in Extended St̲orage (IAEA CRP)
BWR	Boiling Water Reactor
CANDU	**Can**adian **D**euterium-**U**ranium Reactor
CC	Concrete Canister
CLAB	Central Interim Storage Facility for Spent Nuclear Fuel (Sweden)
CRP	Co-ordinated Research Programme
Crud	A deposit on Fuel Assembly surface (in SPAR context)
DSC	Dry Storage Container (Canada)
FA	Fuel Assembly
FR	Fuel Rod
FSM	Field Signature Method
FTU	Formazin Turbidity Unit
HLW	High Level Waste
HTGR	High Temperature Gas-cooled Reactor
ISFSI	Independent Spent Fuel Storage Installation
LWR	Light Water Reactor
MAGNOX	Magnesium No Oxidation (Magnesium Alloy Cladding, UK)
MEB	Multi Element Bottle (UK)
MOX	Mixed Oxide Fuel
MPC	Multi-purpose Canister
MVDS	Modular Vault Dry Storage
NGS	Nuclear Generating Station
NPP	Nuclear Power Plant
NRC	Nuclear Regulatory Commission (USA)
PWR	Pressurized Water Reactor
RBMK	Russian Type of Graphite Modulated Water-cooled Reactor (Reaktor Bolshoy Moshchnosti Kanalniy)
SF (A)	Spent Fuel (Assembly)
SNF	Spent Nuclear Fuel
SS	Stainless Steel
TAD	Transportation, Aging and Disposal
t HM	Tonnes of Heavy Metal
THORP	**Th**ermal **O**xide Fuel R̲eprocessing **P**lant (Sellafield, UK)
WWER	Russian Type of PWR (**W**odo-**W**odyanoi **E**nergetichecki **R**eactor)
Zr1Nb	Zirconium - Niobium alloy fuel cladding (WWER)
ZRA	Zero Resistance Ammetry
Zry	Zircaloy

ANNEX

REPORTS ON RESEARCH PROJECTS WITHIN THE SPAR-II CRP

TITLE: **Determinations of the temperature of terminal solid solubility in dissolution and precipitation of hydrogen/deuterium in irradiated Zircaloy-4**

COUNTRY: **Argentina** CHIEF SCIENTIFIC INVESTIGATOR: **P. Vizcaíno**

AGREEMENT NUMBER: **14377**

COMPANY: **CNEA-CONICET, Centro Atómico Ezeiza**

BACKGROUND:

The proposed plan is an approach to the metallurgical consequences of the high neutron fluencies (10^{22} n/cm^2) on the hydrogen behavior in zirconium based alloys, based on the significance of the microstructural behavior of the high burn up fuel claddings during the dry storage period. The studies are focused on Zircaloy-4, concerning to two processes:

- Neutron irradiation damage
- Hydrogen pick up

The Zircaloy-4 was taken from cooling channels of the PHWR Atucha 1. These components remained more than 10 years in service, reaching neutron fluencies up to 10^{22} n/cm^2.

In the last recent years, measurements of the hydride dissolution temperatures have shown that hydrogen solubility is affected by the neutron irradiation, increasing it respect to the unirradiated Zircaloy solubility. In addition, in this material the amorphization/dissolution of the second phase particles (SPPs) was observed, being proposed an interaction between the hydrogen atoms, the SPPs and the irradiation defects as a possible explanation of the observed behavior. For the present case, attention will be focused on the hydride precipitation process, since it is strongly related with delay hydrogen cracking initiation, a problem of direct concern for the dry storage. The goal of the present proposal is to make an approach to the source of the observed effect, applying several specific techniques as differential scanning calorimetry (DSC), high resolution x-ray diffraction and transmission electron microscopy.

OBJECTIVES

The objectives can be divided as follows:

- Determination of the temperatures of terminal solid solubility in dissolution (TTSSd) and in precipitation (TTSSp) in high fluency irradiated Zircaloy-4, reproducing the temperatures at which the Zircaloy fuel claddings remain during dry storage by an annealing program during the DSC experiments.
- Observations by optical and transmission electron microscopy of the hydride distribution before (as received material) and after high temperature annealings, trying to get deepener on the cause of the observed effects.

RESEARCH APPROACH

As a brief summary of the results, the following points can be underlined:

(1) Considering only the annealing effect of the isothermal treatments made at 350°C and 400°C, and discarding conservatively the dissolution temperature determined at the first run, it was observed for the whole data set a temperature increase higher than 30°C in TTSSd, reaching values of 50°C for a few cases. This increase is higher if the annealings at 500°C and 600°C are considered. If these raise in TTSSd are converted in [H], a hydrogen increase of 50% respect to the calculated from TTSSd measured at the 2nd run is observed.

(2) The precipitation temperature seems to be affected by two processes: radiation damage recovery and the annihilation of the nucleation sites due to the maximum temperature and the time remained at this temperature in each DSC run. As both effects take place simultaneously, the precipitation behavior less regular than the dissolution and the TTSSp values are less accurate. If only the isothermal annealings at 350°C and 400°C are considered, increases of from 10°C to 35°C were observed. If the annealings higher temperatures are included (500°C and 600°C), all the samples increased the precipitation temperature between 25°C to 50°C.

Finally, the analysis of the DSC curves and observations by optical and transmission electron microscopy added to previous x-ray diffraction results indicate that the observed increase in the solubility temperatures are due to a wide distribution of hydride sizes, with a preponderancy of small sizes in the material as received, affecting the temperatures at which the DSC signal evidences the end of dissolution or the start of precipitation. These small hydrides disappear gradually after the annealings as the defect density decreases, tending the dissolution and precipitation temperatures to the unirradiated values.

TITLE: **Behaviour of power and research reactor fuel in wet and dry storage**

COUNTRY: **Canada**

CHIEF SCIENTIFIC INVESTIGATOR: **J. Freire-Canosa**

AGREEMENT NUMBER: **13077**

COMPANY: **Nuclear Waste Management Organization**

BACKGROUND:

Canada has developed extensive experience in both wet and dry storage of CANDU fuel. Fuel has been stored in water pools at CANDU reactor sites for approximately 45 years, and in dry storage facilities for a large part of the past decade. Currently, Canada has 38 450 t U of spent fuel in storage, of which 8850 t U are in dry storage.

In June 2007, the Government of Canada selected the Adaptive Phased Management (APM) approach, recommended by the Nuclear Waste Management Organization (NWMO), for the long-term management of Canada's nuclear-fuel waste.

OBJECTIVES

The Canadian utilities and AECL are conducting development work in extended storage systems as well as research on fuel behaviour under storage conditions. Both activities have as ultimate objective to establish a technical basis for assuring the safety of long-term fuel storage.

RESEARCH APPROACH:

The Canadian contribution to SPAR-II focused on three main areas:

(1) Development of the CANDU Fuel Bundle Stress Model

(2) Development of an Apparatus and Methodology to test the susceptibility of CANDU fuel bundle endplate/endcap welds to Delayed Hydride Cracking (DHC)

(3) Temperature Monitoring of a Dry Storage Container (DSC-812)

A stress model using FEM for CANDU fuel was developed using the computational platform ANSYS to predict stress fields in a bundle and stress intensity factors at the endcap/endplate welds for fuel stored in dry storage. Predictions from this modelling can be further used to assess the susceptibility of CANDU fuel to DHC.

The models were validated against test results obtained from three series of 'pull tests' on a fuel element from a CANDU fuel bundle and recording the bundle displacements at key locations including the bundle endplates. A comparison of model predictions with the test results indicated that the models predict the general trends well but overestimate the fuel element displacements and underestimate the endplates displacements. However, for loads relevant to dry storage and transportation of the spent fuel, the model predictions appear adequate. Further tests and simulations are planned to gain a better understanding of the observed discrepancies between the model and the bundle tests.

A test apparatus was also developed and tested with non-irradiated CANDU fuel bundle endcap/endplate welds. Test procedures for determining initiating stress intensity factors for Delayed Hydride Cracking (KIH) and delayed hydride cracking velocities (DHCV) were established and validated.

Four of five samples tested at 150°C in KIH tests, had KIH values ranging from 7.6 to 7.9 MPa $m^{1/2}$. The other sample had a KIH value of 13.6 MPa $m^{1/2}$, probably due to a difference in weld morphology. In DHCV tests at 150°C, the cracking velocities of the endcap/endplate welds were found to increase after heat tinting at about 200°C. The DHCV values for these tests ranged from 5.7 x 10-10 m/s to 2.1 x 10-9 m/s prior to heat tinting and from 1.3 x 10-9 m/s to 5.5 x 10-9 m/s after the heat tinting. The results suggest that DHC will not be operative in CANDU fuel during dry storage.

A Dry Storage Container (DSC-812) was instrumented with 32 thermocouples to monitor temperatures during its manufacturing, processing and storage when loaded with fuel. The DSC was loaded with near 16 year old CANDU Bruce fuel on June 15, 2007. Results from monitoring during the storage period indicated that the DSC had reached quasi-steady state during the storage period with maximum temperature in the inner liner of about 55°C when the outside ambient air temperature was about 30°C.

TITLE: **Materials in the environment of the fuel in dry storage**

COUNTRY: **France** CHIEF SCIENTIFIC INVESTIGATOR: **H. Issard**

AGREEMENT NUMBER: **13504**

COMPANY: **TN International (Cogema Logistics)**

BACKGROUND:

Spent nuclear fuel has been stored safely in pools or dry systems in over 30 countries. The majority of IAEA Member States have not yet decided upon the ultimate disposition of their spent nuclear fuel: reprocessing or direct disposal. Interim storage is the current solution for these countries. For developing the technological knowledge data base, a continuation of the IAEA's spent fuel storage performance assessment was achieved.

OBJECTIVE:

Investigate the dry storage systems and gather basic fuel behaviour assessment Gather data on dry storage environment and cask materials Evaluate long term behaviour of cask materials

RESEARCH APPROACH:

To improve the knowledge data base on dry storage systems, it is necessary to collect the available existing experience and technologies: wet technologies (pools) or dry technologies (metal casks, concrete casks, vaults).

Concerning fuel degradation, information is collected from experiments and evaluations done by CEA in France, in order to evaluate fuel integrity.

Experience on long term behaviour of dry storage materials and equipments can provide information about spent fuel environment during storage.

Methods and experience for drying spent fuel before storage in casks are considered and research concerning the long term behaviour of metallic seals is performed.

RESULTS:

In France spent fuel is stored in wet conditions (pools) before reprocessing. The wet conditions are less severe for the spent fuel performance than dry conditions since the temperatures are lower.

As package designer, TN international has studied also dry conditions and collected data on cask components and spent fuel behaviour at different temperatures.

Long term tests to evaluate thermal resistance at service temperatures on package components such as neutron shielding at 150°C or metallic seals at 250°C – 20.000 hours have provided substantial evidence showing that the safety functions of these components are kept during the storage period of 40 years. The leak-tightness and shielding performance of the dry storage casks are maintained during the whole period and it has been shown that the thermal condition of spent fuel depends only on the residual heat coming from radioactive materials. These results contribute to the demonstration of spent fuel performance in storage.

TITLE: **Dry storage assessment of LWR fuel in Germany**

COUNTRY: **Germany**

CHIEF SCIENTIFIC INVESTIGATOR: **W. Goll**

AGREEMENT NUMBER: **13079**

COMPANY: **AREVA NP GmbH**

BACKGROUND:

Germany's revised energy act, dated 2002, prohibits the shipment of spent nuclear fuel to reprocessing plants and restricts its disposal to a final repository. To comply with this law and to ensure further nuclear plant operation, the reactor operators had to construct on-site facilities for dry cask storage, to keep spent fuel assemblies for 40 years until a final repository is available. Twelve facilities went into operation during the last years. The amount of spent fuel in store is continuously increasing and has reached a level of about 1700 t HM by end of 2007. The central sites Ahaus and Gorleben remain in operation but shall be used for special purposes in future.

OBJECTIVE:

- Review of main features of facilities with an emphasis on associated monitoring.
- Review of degradation mechanisms in the context of fuel types and design (PWR, BWR, UO2, MOX) relative to fuel burn-up, structural materials and long term behaviour.

RESEARCH APPROACH:

Licensing process and installation of the new interim storage facilities as well as the transfer of spent fuel have been followed. Fuel design and burn-up developments have been assessed with regard to implications for dry storage integrity. Given a still growing discharge burn–up, long term tests on highly burnt PWR and BWR fuel rods were launched to analyse the integrity behaviour of the fuel rod cladding under demanding dry storage conditions.

RESULTS:

In Germany, all new fuel designs will be assessed with regard to cladding integrity according to the German guidance for spent fuel in dry storage casks. Being aware that in case of very high burn-ups or high duty MOX utilization, existing cask licensing limits may be challenged, further R&D activities were started on commercial BWR and PWR cladding materials with burnups between 55 and 80 GWd/t U. These long-term thermal creep tests have been performed at about 400°C and circumferential stresses of 130 to 150 MPa over 2 to 3 years and are especially intended to provide information on cladding integrity and on hydrogen effects under thermal cycling conditions. As the measurements did not proceed as expected, some samples still have to be tested and caught up with destructive analysis. Only preliminary results are available to date. They indicate that, even at very high burn-ups under thermal cycling conditions, the German engineering limits of 120 MPa circumferential stress and 1% strain are not challenged.

The thermal creep tests on irradiated materials shall also provide data to improve the creep model used for integrity assessment. Up to now, the creep modelling has been based on non-irradiated material laws. First results are positive and it is expected that new creep laws from irradiated materials will considerably reduce the conservatism of the currently used enveloping material concept.

TITLE: **Assessment of spent WWER-440 fuel performance under long-term storage conditions**

COUNTRY: **Hungary**

CHIEF SCIENTIFIC INVESTIGATOR: **F. Takáts**

CONTRACT NUMBER: **13080**

COMPANY: **TS Enercon Kft**.

BACKGROUND:

Paks Nuclear Power Plant is the only NPP in Hungary. It has four WWER-440 type reactor units. The fresh fuel is imported from Russia so far. The spent fuel assemblies were shipped back to Russia until 1997 after about 6 years cooling at the plant. A dry storage facility (MVDS type) has been constructed and is operational since then. By 1 January 2008, there were 5107 assemblies in dry storage.

OBJECTIVE:

- Wet AR storage of spent fuel from the NPP Paks:
  - Measurements of conditions for spent fuel storage in the at-reactor (AR) storage pools of Paks NPP (physical and chemical characteristics of pool water, corrosion product data)
  - Measurements and visual control of storage pool component characteristics
  - Evaluation of storage characteristics and conditions with respect to long-term stability (corrosion of fuel cladding, construction materials).
- Dry AFR storage at Paks NPP:
  - Calculation and measurement of spent fuel conditions during the transfer from the storage pool to the modular vault dry storage (MVDS) on the site
  - Calculation and measurement of spent fuel conditions during the preparation of fuel for dry storage (drying process), such as crud release, activity build-up
  - Measurement of spent fuel conditions during the long-term dry storage, activity data in the storage tubes and amount of crud.

RESEARCH APPROACH:

Aiming to evaluate physical and chemical conditions in the storage pools the following parameters are monitored: temperature (^{o}C), pH, optical transparency (%), chemical composition (Cl, H_3BO_3), isotope-specific activity (Bq/kg).

The overall limitations ($t < 70$ ^{o}C in case of refuelling operations, $< 60^{o}$C, in case of storage; $H_3BO_3 > 12$ g/kg) are continuously controlled. Chemical conditions are measured and corrected if necessary minimum daily during refuelling operations, and weekly during storage, except the boron concentration monitored continuously.

As routine measurements the following activity data are measured:

- Corrosion products (^{51}Cr, ^{54}Mn, ^{59}Fe, ^{58}Co, ^{60}Co, ^{110m}Ag)
- Volatiles (^{131}I, ^{132}I, ^{133}I, ^{134}I, ^{135}I,)
- Non-volatile components (^{137}Cs, ^{134}Cs, ^{138}Cs, ^{139}Ba, ^{140}Ba, ^{91}Sr, ^{92}Sr, ^{85}Rb, ^{88}Rb, ^{89}Rb)
- Other components (^{187}W, ^{56}Mn, ^{99}Mo, ^{122}Sb, ^{124}Sb, ^{24}Na, ^{42}K, ^{95}Nb, ^{95}Zr, ^{97}Zr, ^{140}La).

The measurements of activity are carried out — as a rule — once in month, during refuelling measurements they are done more frequently.

RESULTS

The conclusions drawn are similar to those of the earlier research timeframe, namely:

- On the basis of the measured chemical characteristics, the water quality remains in stable condition. The pH values vary between 4.5 -6.2, the lowest value of optical transparency is 95.4 %. The activity levels can be managed safely using the pool water purification system.
- The main sources of activity in the storage pools are surface and water contaminants. Corrosion during storage period is negligible.
- Dry storage experience is good; temperature of the stored fuel is low, as confirmed by the benchmarked calculations. There were some storage tubes reopened and no unexpected observations were made.

Preparations are made for dry storage of damaged spent fuel and for fuel with surface deposits.

TITLE: **Long term integrity of spent fuel and construction materials for dry storage facilities**

COUNTRY: **Japan**

CHIEF SCIENTIFIC INVESTIGATOR: **T. Saegusa**

AGREEMENT NUMBER: **13082**

COMPANY: **CRIEPI**

BACKGROUND:

In Japan, two dry storage facilities at reactor sites have already been operating since 1995 and 2002, respectively. Additionally, a large scale dry storage facility away from reactor sites is under safety examination for license near the coast and desired to start its operation in 2010. Its final storage capacity is 5,000tU. It is therefore necessary to obtain and evaluate the related data on integrity of spent fuels loaded into and construction materials of casks during long term dry storage.

OBJECTIVES:

- Spent fuel rod
    - To evaluate hydrogen migration along axial fuel direction on irradiated claddings stored for twenty years in air
    - To evaluate pellet oxidation behaviour for high burn-up UO_2 fuels
- Construction materials for dry storage facilities
    - To evaluate long term reliability of welded stainless steel canister under stress corrosion cracking (SCC) environment
        - ♦ To evaluate long term integrity of concrete cask under carbonation and salt attack environment
    - To evaluate integrity of sealability of metal gasket under long term storage and short term accidental impact force

RESEARCH APPROCH:

Two PWR-UO_2 spent fuels stored for 20 years under dry condition, whose burn-ups are 58 and 31 GWd/t U, respectively, were tested to obtain data related to hydrogen migration under temperature gradient along cladding axial direction, and the hydrogen axial migration was estimated by using these experimental data. The pellet oxidation experiments were carried out on irradiated UO_2 fuels whose burnups are 50 GWd/t U and 65 GWd/t U. The metal and concrete casks were used to verify the integrity of construction materials. The experimental and computational analysis items; for the full scale metal cask: (1) drop test without impact limiters, (2) airplane crash test, (3) long-term sealability test of lid structure. For the full scale concrete cask: (1) canister drop test, (2) seismic test, (3) SCC evaluation test.

RESEARCH RESULTS:

- Spent fuel rod

  - There was no significant change in the calculated axial hydrogen profile using heat of transport, diffusion coefficient and solubility limit of irradiated Zircaloy claddings after 40 years of storage under helium atmosphere.
  - At temperature of 300 °C, there was not burn-up dependence in the oxidation behaviour. But at 350 °C, the oxidation rate in the sample with burn-up of 65 GWd/t U was higher than that with 50 GWd/t U beyond the O/M of 2.4. The pellet microstructure may affect oxidation behaviour.

- Construction materials for dry storage facilities

  - For drop tests (1m horizontal and 1m rotational) of the full scale metal cask, the leak rate value from the secondary lid increased by three or four orders of magnitude during the impact immediately. The leak rate recovered by two orders of magnitude 20 minutes later after the drop test. Moreover, the change of the inner pressure between two lids was negligible.

  - For aircraft crash test on the metal cask using the geometry scale factor of 1/2.5, it seems that the loss of the inner pressure of the cask cavity may be avoided in the impact event with the horizontal orientation even if the severe impact load was applied on to the metal cask due to aircraft engine crash.
  - Long-term containment of the secondary metal gasket in the cask lid structures of the full-scale models had been measured for more than 17 years under a constant temperature of 160 °C at the primary lid gasket. Until the end of March in 2008, the sealability has been well maintained.
  - For the canister, helium leak tests were performed before and after the 1m horizontal and 6m vertical drop tests. Measured leakage rates in both tests showed the integrity of sealability at lids and canister shell. Namely, all values were under 1.0×10^{-9} Pa • m^3/s.
  - For seismic tests using a full scale concrete cask, it was found that the tip-over of the full scale cask did not occur, but the special attention should be paid for the sliding or jumping behaviour of the cask under the strong earthquake motion.
  - For concrete cask, threshold chloride density for emerging rust, chloride deposition velocity on the metal surface and crack growth rate of candidate canister materials were measured.

TITLE: **Evaluation of Thermal Creep and Hydride Re-orientation Properties of High Burnup Spent Fuel Cladding under Long Term Dry storage**

COUNTRY: **Japan**

CHIEF SCIENTIFIC INVESTIGATOR**: K. Kamimura**

AGREEMENT NUMBER: **13081** COMPANY**: JNES**

BACKGROUND

In Japan, spent fuels will be reprocessed as recyclable energy source at a reprocessing plant. The first commercial plant is under-constructing and will start operation in 2008. It is necessary that spent fuels should be stored in the independent interim storage facilities (ISF) until reprocessing. Utilities plan the operation of the first ISF in 2010. JNES has a mission to support the safety body by researching the data of technical standard and regulation. Investigating of spent fuel integrity during long term dry storage is one of them.

OBJECTIVES

(1) Evaluation of the effects of material design changes on creep properties of high burnup spent fuel cladding
(2) Evaluation of the effects of alloy elements and texture of irradiated Zircaloy on hydride re-orientation properties and the effects of radial hydrides on cladding mechanical properties
(3) Evaluation of the effects of temperature on irradiation hardening recovery.

RESEARCH APPROCH

(1) Creep and Creep Rupture Test
    - Material: 50GWd/t type BWR and 48GWd/t type PWR fuel cladding tube (Zry-2 and -4) 55GWd/t type BWR and PWR fuel cladding tube (Zry-2, MDA and ZIRLO)
    - Temperature: 330~420 °C
    - Time: up to 11,000 h
    - Hoop Stress: 30~300MPa

(2) Hydride Reorientation Effect Evaluation Test
    - Material: 40, 50GWd/t type BWR and 39, 48GWd/t type PWR fuel cladding tube (Zry-2 and -4)
    - 55GWd/t type BWR and PWR fuel cladding tube (Zry-2, MDA and ZIRLO)
    - Temperature: 250~400 °C
    - Hoop Stress: 16~130MPa

(3) Irradiation Hardening Recovery Test
    - Material: 50GWd/t type BWR and 48GWd/t type PWR fuel cladding tube (Zry-2 and -4)
    - Temperature: 330~420 °C
    - Time: up to 11 000 h

RESEARCH RESULTS

(1) Creep and Creep Rupture Test

- ✓ The thermal creep test for irradiated Zry-2 and Zry-4 cladding tube were performed, and the creep equations for each material were formulated from the creep test results.
- ✓ Irradiated materials suppress creep compared to unirradiated material, but degree of suppression depends on temperature and stress conditions.
- ✓ No effects of hydride reorientation in radial direction have been observed on creep rate.
- ✓ The threshold strain of transition to tertiary creep is likely to be more than 10 % for unirradiated cladding and 1% for irradiated cladding.

(2) Hydride Reorientation Effect Evaluation Test

- ✓ *Irradiated BWR Zry-2 cladding*

  The hydride reorientation to radial direction occurred in relatively low HRT hoop stress, such as less than 70MPa. The increase of reorientation with hoop stress was not monotonic for the specimens in which a part of hydrides remained precipitated at HRT temperature, such as the case for 50GWd/t type cladding at 300ºC HRT.

  The ductility of the specimens after 400℃, 0MPa, 30ºC/h HRT increased in ring compression testing at room temperature compared to no HRT (as-irradiated) specimens, and it is indicated that the recovery of irradiation damage at 400ºC annealing occurred and affected the ductility of the irradiated Zry-2 cladding.
- ✓ *Irradiated PWR Zry-4 and improved Zirconium alloy cladding*

  Little increase of the radial hydride ratio occurred in HRT 100MPa, 340ºC or less. On the other hand, the amount and the length of the hydride in the mid wall area of cladding depended on the temperature and cooling rate in HRT due to the hydrogen migration from the hydride rim area.

  It is deduced that the ductility in ring compression deformation was affected by the orientation, amount, and length of hydride in the mid wall area.

(3) Irradiation Hardening Recovery Test

- ✓ The irradiation hardening recovery was observed after 330 °C, 8000h or more annealing for both irradiated Zry-2 and Zry-4 cladding. In addition, recovery data at 300°C annealing were also acquired. No recovery in hardness was observed for PWR 48GWd/t type Zry-4 cladding. As for BWR 50GWd/t type Zry-2, a slight decrease in hardness was observed after 300 °C, 5000h annealing. Different temperature in reactor core between BWR and PWR may affect the recovery behaviour.

TITLE: **Assessment of spent WWER-440 fuel performance under long-term storage conditions**

COUNTRY: **Slovakia**

CHIEF SCIENTIFIC INVESTIGATOR: **Ľ.Kupča**

CONTRACT NUMBER: **13084** COMPANY: **VUJE Inc.**

BACKGROUND:

In the Slovak Republic are under operation 6 units (4 in the Jaslovské Bohunice site, and 2 in the Mochovce), 2 units are under construction in Mochovce site. All units are WWER-440 type. The fresh fuel is imported from the Russian Federation. The spent fuel assemblies are stored in wet conditions in Bohunice Interim Storage Spent Fuel Facility (SFIS). By 15 July 2008, there were 8413 assemblies in SFIS.

OBJECTIVE:

(1) Wet AR storage of spent fuel from the NPP Bohunice and Mochovce:

- Surveillance of conditions for spent fuel storage in the at-reactor (AR) storage pools of both NPP`s (characteristics of pool water, corrosion product data)
- Visual control of storage pool components
- Evaluation of storage conditions with respect to long-term stability (corrosion of fuel cladding, structural materials).

(2) Wet SFIS storage at Bohunice:

- Measurement of spent fuel conditions during the long-term wet storage, activity data in the storage casks and amount of crud
- Surveillance program for SFIS structural materials.

RESEARCH APPROACH:

Following physical and chemical parameters in the storage pools are monitored:

Basic parameters of the cooling water in storage pools

Parameter	Units	Limiting	Typical values
pH	–	5.5 ÷ 8.0	5.8 ÷ 6.5
Cl-	$mg.kg^{-1}$	< 0,1	< 0,02
electrical conductivity	$\mu S.cm^{-1}$	< 3	1 ÷ 1,5
suspended particles	$mg.kg^{-1}$	< 0,5	< 0,4
3H (T)	$Bq.m^{-3}$	$< 3,7.10^{9}$	10^{5}
total activity	$Bq.m^{-3}$	$< 4,1.10^{7}$	$10^{2} \div 10^{3}$
temperature	°C	< 50	20 ÷ 50

RESULTS

- On the basis of the measured chemical characteristics, the water quality remains in stable condition as in the table above due to the periodical cleaning.
- From the analyses of surveillance specimen follows that corrosion of monitored materials during storage period is negligible.
- Extension the storage capacity of SFIS is under evaluation with respect of planned construction of new NPP units.

TITLE: **Spent fuel and materials performance in wet and dry storage**

COUNTRY: **Spain**

CHIEF SCIENTIFIC INVESTIGATOR: **P. Zuloaga**

AGREEMENT NUMBER: 13085

COMPANY: **ENRESA**

BACKGROUND:

According to the 6th General Radioactive Waste Plan, spent fuel in Spain shall have to be gathered in a Centralised Temporary Storage (CTS) during some decades in order to have time for a decision concerning its final fate: direct disposal at a geological repository or partitioning and transmutation if technology opens this possibility when the decision will be taken, expected in 2050. The CTS technology has already been chosen as a vault type building based in spent fuel dry storage. To support the use of this technology, a number of programmes have been completed or are still in progress, mostly concerned about high burnup fuel issues and new cladding materials. These programmes are directly managed by ENRESA alone or in joint venture with other parties, at a national and international level. Apart from that, there are contacts with other countries organisms who share similar interests with Spanish ones.

OBJECTIVE:

- Review of spent fuel data relevant for future storage in Spain

- Perform destructive and non-destructive examinations on irradiated and non-irradiated fuel rods relevant to Spanish spent fuel management.

RESEARCH APPROACH**:**

Among the programmes initiated in the last years (finished or still in progress) one may highlight the following ones:

- Isotopic measurements on high burnup fuels: up to 75 GWd/t U PWR and 53 GWd/t U BWR peak values.
- Mechanical tests on high burnup PWR – ZIRLO cladding and BWR – Zry2 cladding samples, including axial tensile tests, ring tensile tests, ring compression tests and creep tests. Hydrides behaviour is also measured.
- Mechanical tests on unirradiated ZIRLO rods
- Hydrides behaviour modelling

RESULTS

Spent fuel management is an important issue for fuel cycle costs. Any method for optimizing the amount of spent fuel to store and transport produces increasing profits to nuclear industry. The spent fuel volume stored or transported in a given system is limited by several factors, i.e. reactivity, heat generation and radiation doses. The methodologies used for safety analysis of these systems include several conservatisms caused by the limited knowledge of several aspects of spent fuel behaviour. Moreover, many of the limits in the regulatory guidance related to spent fuel have not followed the evolution of increasing fuel burnup discharge and designs. An example of this is the criticality analysis with burnup credit for reactivity determination based on isotopic calculations. These calculations involve high uncertainties. A reduction of uncertainties, and the associated conservatism, can be obtained through experimental measurements, which help closing knowledge gaps and validating calculation codes. ENRESA and her partners have obtained isotopic measurements on PWR – high burnup and high enrichment samples for isotopes representative of shielding, gas production, reactivity, residual heating and burnup determination. A different approach has been followed concerning BWR samples. Although the BWR-samples' burnup is lower, the aim is to highlight the voids' effect and the different neutron spectra of a BWR compared to a PWR. BWR measurements are still in progress.

Five high burnup PWR-ZIRLO samples were creep tested and characterized to investigate the behaviour of advanced cladding materials under dry storage conditions. Creep test matrix included 360-400 ºC and 160-220 MPa hoop stress conditions. Moreover, hydride reorientation was investigated imposing specific cool-down terms. The results have been consistent with the expected behaviour from reference CWSR material, Zry4. The higher hoop stress and temperature, the greater creep deformation. The samples kept significant ductility during tests. Only one sample had a leak after 17% engineering strain of the tube. Metallographic tests done afterwards showed significant hydride reorientation, but without any specimen failure observed during cool-down. Hardness measurements showed partial recovery in some of them. Some samples were also mechanically tested and were useful to evaluate high burnup cladding properties.

The BWR-Zry2 samples' testing is in its early beginning. No results can be presented yet. A confirmation of better creep behaviour is expected, as it is generally observed comparing RX material to CWSR one.

Other tests on unirradiated ZIRLO rods have been performed to get the mechanical properties and behaviour of this type of cladding, at ambient temperatures and at other temperatures representative of in-pile or storage conditions. A cohesive fracture model has been chosen for its modelling. A hydrogen charging method has been added to show the effect of hydrides. Moreover, by carefully selecting stresses and temperatures, a more or less degree of hydride reorientation can be obtained. Mechanical tests include axial tensile, ring tensile and ring compression tests. The results have let the researchers develop an iterative model that is thought to be checked on irradiated material cases.

Finally, a hydride migration modelling has been developed and tested against some example cases, taken from literature and from experimental cases. In particular, irradiation and temporary storage has been modelled. Another important target of this modelling includes hydrogen migration to colder areas, such as pellet-pellet interfaces or a hydrogen blister caused by oxide layer removal. A 2-D model is being developed. It is expected to be improved to a 3-D model and to be connected to a mechanical model to include hydride reorientation.

TITLE: **Corrosion inhibition studies in support of the long term storage of AGR fuel**

COUNTRY: **UK**

CHIEF SCIENTIFIC INVESTIGATOR: **P. Standring**

AGREEMENT NUMBER: **13086**

COMPANY: **Sellafield Limited**

BACKGROUND:

Thorp Receipt and Storage (at Sellafield, UK) is currently being investigated as a bridging solution for the storage of AGR fuel pending the out-come of a national review into spent fuel management.

AGR spent fuel is known to be susceptible to corrosion through inter-granular attack. To avoid this, the chosen storage regime for AGR fuel is sodium hydroxide dosed pond water to pH 11.4; now 22 years of operating experience. The conversion of TR&S will require a phased transition. During this transition sodium hydroxide cannot be used due to materials compatibility issues. Alternative corrosion inhibitors have been investigated as an interim measure and sodium nitrate has been selected as a suitable candidate. The efficiency of sodium nitrate to inhibit propagating inter-granular attack of active AGR materials has yet to be established.

In the longer term sodium hydroxide will be deployed along with a move to a closed loop pond water management system. Given that carbon dioxide is known to be absorbed by sodium hydroxide dosed water and can affect fuel integrity, in the case of Magnox fuel, there is a need to establish its impact on AGR fuel.

OBJECTIVES:

- To establish the impact of carbonate on AGR fuel corrosion
- To establish the efficiency of sodium nitrate to inhibit propagating inter-granular attack of irradiated AGR materials

RESEARCH APPROACH:

The impact of carbonate will be evaluated inactively in the first instance by producing potentio-dynamic polarisation curves. The corrosion specimen will be of the ZRA iga type fabricated from sensitised AGR clad.

The efficiency of sodium nitrate will investigated using zero resistance ammetry (ZRA). ZRA probes will be fabricated from active AGR brace material with 304L being used as a reference electrode. The AGR brace specimens will be first selected from running corrosion test in 2ppm chloride solutions to find the worst case material.

RESEARCH RESULTS:

- Effect of carbonate on AGR fuel corrosion – At low levels, up to 150ppm, there is no effect of carbonate. At high levels, 300 and 500ppm, there is evidence that carbonate acts as a corrosion inhibitor. Based upon considerations of AGR fuel corrosion there would appear to be no need to remove carbonate from pond water on fuel integrity grounds.

- Brace samples from a number of positions along an AGR stringer were fabricated into ZRA sensors and subject to corrosion in chloride (2ppm). From these initial scoping trials the worst case AGR brace material was selected for testing with sodium nitrate.
- Initial testing of AGR brace material with sodium nitrate has been initiated; 4 tanks with 12 samples in each. Corrosion of the ZRA sensors fabricated from AGR brace material were initiated in chloride (2ppm). Tests solutions for each of the tanks were adjusted to 2ppm chloride at 40 and 50ºC and 0.5ppm chloride at 40 and 50ºC. 10ppm sodium nitrate was then dosed to each of the tanks and the time to arrest corrosion established. Initial results suggest that the samples at 2ppm chloride at 50º were passivated within 10 days.

TITLE: **US spent fuel research and experience**

COUNTRY: **USA**

CHIEF SCIENTIFIC INVESTIGATORS: **A. Machiels**

AGREEMENT NUMBER: **13087**

COMPANY: **EPRI and USDOE**

BACKGROUND**:**

The structural performance of high-burnup spent fuel cladding during dry storage and transportation has been the subject of research and evaluation at EPRI for several years. The major issues addressed in this research program have included the following:

- Characterization and development of predictive models for damage mechanisms perceived to be potentially active during dry storage
- Modeling and analysis of deformation processes during long-term dry storage
- Development of cladding failure models and failure criteria, considering cladding material and physical conditions during dry storage and transportation
- Failure analysis, considering end-of-dry-storage conditions, of spent fuel systems subjected to normal and accident conditions of transport, prescribed in Part 71 of Title 10 of the Code of Federal Regulations (10CFR71)

While issues related to dry storage have largely been resolved, transportation issues have not, at least for spent fuel with discharge burnups greater than 45 GWd/MTU. A research program was launched in late 2002 following two NRC-industry meetings held on September 6, 2002 and October 23, 2002. The aim of the research program was to assess the performance of high-burnup spent fuel cladding under normal and accident conditions of transportation, as prescribed by 10CFR71, considering the physical characteristics and mechanical properties of cladding at the end of dry storage.

OBJECTIVE:

To present a synthesis of the information that collectively forms a part of a technical basis intended to facilitate resolution of regulatory issues associated with the transportation of spent nuclear fuel characterized by discharge burnups greater than 45 GWd/MTU.

RESEARCH APPROACH:

The research team reviewed the large body of modeling and analysis work carried out during the past several years and then organized it into three technical areas. The first technical area examines dry storage effects on cladding geometry (due to creep) and hydride morphology in the cladding alloy (due to hydride re-orientation). The second technical area deals with the development of failure models and failure criteria for application to transportation accident analysis. The third technical area applies the above-described developments to the evaluation of fuel rod failures in transportation casks subjected to loading conditions specified in 10CFR71.

RESEARCH RESULTS:

The results of the analytical studies conducted during the past several years indicate that damage to high-burnup spent fuel under prescribed regulatory conditions of dry storage and transportation will not impair its operational management. Dry storage effects on cladding physical and material conditions, including creep-related deformations, have the potential to impact fuel rod performance during transportation. Specifically, cladding resistance to failure under the dynamic loading of transportation accidents depends on fuel-cladding gap size and radial hydride formation, both of which could—to a limited extent—evolve during long-term dry storage. The results of the hypothetical accident analysis indicate that cladding failure would be bi-modal—taking the form of (1) a state of failure initiation at the cladding inside diameter (ID) remaining as part-wall damage, with less than a 2% probability of occurrence and (2) a through-wall failure with a probability of 1E-5. The response analysis under normal conditions of transport shows a large margin against fuel rod failures. The grids and guide tubes, which form the structural elements of the fuel assembly, are predicted to remain structurally competent. As a result, the geometric form of the spent fuel assemblies will not be substantially altered.

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Consultants Meeting

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