CANDU Fuel Long-Term Storage and Used-Fuel Integrity

Z. Lovasic, P. Gierszewski
Ontario Power Generation

ABSTRACT

Technologies for long-term dry storage in helium or air have been developed for used CANDU fuel. Used fuel dry storage facilities are presently in operation or under construction at all Canadian nuclear power stations.

The evaluation of the long-term integrity of stored used fuel started in 1977. Direct examination of fuel stored under water for up to 27 years has indicated that CANDU fuel, defected or not, could be safely stored. The Easily Retrievable Basket and Controlled Environment Experiments have provided information about potential impacts of aging mechanisms during used fuel storage in air for times up to 20 years.

The used fuel aging investigation focused on four mechanisms that are believed could have long term impact on used fuel integrity: Stress Corrosion Cracking (SCC), UO2 oxidation, creep and Hydrogen Assisted Cracking (HAC). As a result it was concluded that under normal dry storage conditions it is unlikely that CANDU fuel will suffer significant degradation during a period of 100 years. Additional investigation is still necessary in the areas where there is a higher uncertainty in the prediction of used fuel condition and for the degradation processes that are potentially more aggressive. Additional work is also necessary to evaluate the possible effects of abnormal/accident conditions on the integrity of fuel.

In 2004 a new program on used fuel integrity was started. The outlines of this program will be discussed. In the first phase of this project, the focus is on a review of the used fuel characteristics at the beginning of dry storage, and on studies of likely stresses that the fuel will be exposed to during handling and transportation after dry storage. In addition, preservation of relevant information (form and content) about the used fuel during a 100 year period is also being studied. In the first phase most of the investigation is carried out by Nuclear Safety Solutions (NSS). After the first phase additional examination and testing of used fuel is planned.

1.0 INTRODUCTION

This paper describes the approach to and status of the OPG Used Fuel Integrity (UFI) investigation project in 2005. The project started in 2004 and is planned to last five years. The project is also a part of the IAEA Coordinated Research Project (CRP) in "Spent fuel performance assessment and research (SPAR II)".

2.0 CANDU FUEL DESIGN AND LIFE CYCLE CONDITIONS RELATED TO USED FUEL INTEGRITY

CANDU fuel bundle design has evolved since the first developments of the CANDU reactor technology. The first fuel bundles used at the Nuclear Power Demonstration (NPD) reactor near Chalk River with natural uranium contained 7 fuel elements. The fuel bundles used at Douglas Point nuclear station consisted of 19 fuel elements. Current CANDU operating stations in Ontario use two types of fuel bundle design. The Pickering station uses a 28-element bundle design, while the Bruce and Darlington nuclear stations use a 37-element bundle design. About 30 % of the fuel bundles currently used is slightly longer (approximately 1 cm). Low void reactivity fuel is also being considered for future use.

A characteristic of all the fuel bundle designs is that fuel elements are held together by means of welds attaching the end-caps of each fuel element to two Zircaloy-4 end plates. This type of assembly, unique to CANDU fuel, makes each fuel element an active component of the bundle structure and mechanically constrains each Zircaloy tube via a rigid attachment of each end cap to an end plate.

The typical burnup of CANDU fuel is about 220 MWh/kgU, with a most fuel between 120 and 320 MWh/kg U. The coolant temperature range is 260 to 300 ^{o}C at a pressure of 10 MPa. A comparison of some key characteristics of CANDU and LWR nuclear fuel is shown in Table 1.

Table 1: Comparison of CANDU and LWR UO_2 Fuel Properties (McMurry et al., 2003)

Fuel Property	CANDU Fuel	LWR Fuel
Uranium processing	Natural UO_2	Enriched UO_2 containing 1-5% U-235
UO_2 density (% of theoretical)	97%	94 - 95 %
Oxygen/Uranium ratio	2.001	2.001
Grain size	5 - 10 μm	2 - 4 μm
Fuel element per bundle	28 - 37	50 – 300
Fuel element length	0.5 m	3 - 5 m
Fuel pellet diameter	12 - 14 mm	8.5 - 11 mm
Burnup range	120 - 320 MWh/kg U	190 - 960 MWh/kgU
Average burnup	220 MWh/kg U	625 MWh/kg U
Linear power range	20 - 55 kW/m	15 -25 kW/m
Centre line temperature	800 - 1700 ^{o}C	800 - 1200 ^{o}C

The fuel cladding is Zircaloy. On the inside of the fuel cladding is the CANLUB layer, a graphite-based lubricant that provides an advantage during fuel manufacturing, and also improves the fuel cladding lifetime by adsorbing some elements like iodine that may act as a catalyst for stress corrosion cracking. During reactor operation, the fuel cladding receives a significant radiation dose and may be subjected to chemical corrosion processes. Typically, CANDU fuel cladding is exposed to high-energy neutron fluence in a range from 6 to 10 x 10^{24} n/m^2. The general effect of fast neutron irradiation on the mechanical properties of Zircaloy is increased brittleness.

The most important chemical effect on a fuel bundle is the incorporation of hydrogen and deuterium in the Zircaloy cladding. The hydrogen content in irradiated fuel results in the precipitation of hydrides, which may in certain cases result in cladding failures during reactor operation.

Following their in-reactor service life, the fuel bundles are removed from the reactor and transferred dry to the irradiated fuel bay. This transfer normally takes less than three minutes, during which time the fuel bundle temperature rises and the fuel cladding reaches temperatures of between 300 and 400°C depending on the bundle irradiation history. Following this transfer, the fuel is cooled off by contact with the water of the fuel bay.

After about 10 years in the fuel bay, the used fuel is transferred to dry storage containers. At OPG, 384 bundles are loaded into a Dry Storage Container (DSC). This is then vacuum dried, welded and refilled with helium. The peak temperature within the DSC with helium has been estimated as less than 180^{o}C, and probably no more than 150^{o}C for average burnup fuel. The temperature then decreases as the radioactive heat decays, and oscillates seasonally.

The integrity of the used fuel after an extended period of storage depends in part on the fuel conditions at the start of storage. The amount of fission gas in a fuel element, the neutron damage and hydrogen content of the cladding and, in general, most chemical, thermal and mechanical in-reactor damage to the fuel increases with burnup. (The rate of oxidation of UO_2 to U_3O_8 appears to be lower for fuel with higher burnup.) Burnup varies between bundles, and also within the bundle, with the burnup of the outer fuel elements being about 1.4 times higher than that of fuel elements at the centre of the bundle.

The gas pressure in a fuel element and the resulting net cladding pressure range from 0.41 to 2.4 MPa. The associated localized stresses in the individual fuel elements also vary over a substantial range but under storage conditions remain below 80 MPa for most of the fuel. 100 MPa can be considered the absolute maximum localized stress to which the cladding might be subjected at the assumed high cladding temperature of 180^o C.

3.0 QUANTITIES OF USED FUEL FOR DRY STORAGE

In Ontario there are three nuclear generating stations (Pickering, Bruce and Darlington) with a total of 20 CANDU nuclear reactors. All nuclear fuel will be, after a period of wet storage in the Irradiated Fuel Bays, stored in the dry storage facilities at the site where it is generated.

The quantities of used fuel that will be stored in dry storage facilities depend on the assumed reactor lifetime. A recent reference scenario used by the Nuclear Waste Management Organization assumes that there will be about 3 200 000 fuel bundles in dry storage generated by the OPG nuclear reactors, and a total of about 3,600,000 bundles from all sources in Canada by the year 2033 (NWMO web site, Fact Sheet, Nuclear Fuel Waste in Canada).

4.0 USED FUEL LONG TERM MANAGEMENT

In 2002, a Nuclear Waste Management Organization (NWMO) was formed as a result of the Nuclear Fuel Waste Act. The mandate of the NWMO is to investigate long-term approaches for managing Canada's used nuclear fuel, and to implement a recommended approach. The draft recommendation of the NWMO for the long-term management of used fuel is the Adaptive Phased Management Approach, (NWMO web site, 2005). This approach is divided into three phases, with flexibility in moving between the different phases. While a deep geologic repository is proposed in the final phase, the approach includes an extended period of continued dry storage at reactor sites, and possibly at a central site, before the repository.

In 2004, as a part of the used fuel long-term management technology program, OPG initiated a 5-year project on Used Fuel Integrity during storage. The project is an extension of the previous used fuel integrity studies that started in Canada as early as 1977.

OPG used fuel dry storage systems and structures are in service at the Pickering Waste Management Facility (PWMF) and the Western Waste Management Facility (WWMF) at Bruce site. A third dry storage facility is being built within the Darlington site. Used fuel is placed in Dry Storage Containers (DSCs), which are arranged above ground in storage buildings.

The other Canadian nuclear stations at Gentilly and Point Lepreau have their own technology for the used fuel long term dry storage. They use Concrete Canisters (CCs) designed by AECL (CANSTOR system). Concrete Canisters are also used for storing used fuel from the shutdown CANDU reactors - Whiteshell Laboratories research reactor (WR-1), Douglas Point and NPD (Frost 1994).

There are differences in storage conditions in DSCs and CCs that will have to be taken into account for used fuel integrity studies. In DSCs, there is a helium atmosphere while in CCs it is air. Also, in DSCs the fuel bundles are stored horizontally, while in CCs fuel is stored vertically, which has an impact on stresses.

5.0 PREVIOUS STUDIES OF POTENTIAL DEGRADATION MECHANISMS OF CANDU FUEL

The first formal evaluation of the long-term integrity of used fuel during storage was initiated in 1977 by Ontario Hydro and AECL (Frost 1994).

Examination of fuel stored under water for up to 27 years indicated that neither the cladding nor the UO_2 fuel matrix, in any of the undefected elements, had experienced any apparent degradation (Wasywich and Frost, 1991). The cladding of a defected element that had been stored under water for about 21 years also did not experience any apparent deterioration. It was concluded then that CANDU used fuel, defected or not, can be safely stored under water for at least 50 years (Wasywich and Frost, 1991).

In the 1980s, experiments in concrete canisters started at AECL's Whiteshell Laboratories as an investigation of used fuel integrity during dry storage. The concrete canister experiments were known as the Easily Retrievable Basket (ERB) experiment and the Controlled Environment Experiment (CEX-1 and CEX-2) (Frost, 1994). The ERB experiment involved storage in dry air at seasonally varying temperatures. The Controlled Environment Experiment was divided into two phases, one was the storage in dry air at 150 ^{o}C (CEX-1) and the other was the storage in moisture saturated air at 150 ^{o}C (CEX-2).

The ERB experiment involved storage of two undefected used fuel bundles from Pickering nuclear station. CEX-1 and CEX-2 involved storage of four undefected used fuel bundles from Pickering nuclear station and four bundles from Bruce nuclear station (Frost, 1994). In 1986 two fuel elements that had developed a through-wall cladding defect in-reactor were introduced into the program to compare the effects of deliberately defected elements and in-reactor defected elements. The defected and undefected fuel elements are separated in the storage canisters.

Subsequent retrieval of fuel elements from ERB, CEX-1and CEX-2 experiments, as well as their inspection and destructive analysis, have provided a data base for this and future investigations of used fuel integrity. The latest retrieval of elements from ERB and CEX-1 occurred in 1997.

The following aging mechanisms have been investigated:
- cladding creep;
- UO2 oxidation;
- stress corrosion cracking of the Zircaloy bundle;
- hydrogen assisted cracking (or delayed hydride cracking) of the Zircaloy bundle.

5.1 Cladding Creep

Cladding creep in CANDU fuel has been evaluated based on extrapolations, mechanistic models and creep correlations mostly developed for LWR fuel. Inspections and testing of fuel from ERB, CEX-1 and CEX-2 experiments also provided some inputs for the creep evaluation.

The cladding creep models developed to predict behavior of LWR fuel cladding have been applied to the CANDU fuel. The Larson -Miller model and the mechanistic approach with the diffusion controlled cavity growth mechanism used by NRC and DOE, assuming fuel cladding temperature of 180 ^{o}C were applied. The results from all the models have ruled out creep as a risk to fuel integrity over 100-year dry storage time scales.

However, in CANDU fuel, creep strain can result in changes to the bundle geometry. After irradiation, fuel elements are no longer straight and bundles no longer have a straight cylindrical shape; they are slightly bowed, and the end plates are no longer parallel to each other. The deformation induced on a fuel element during irradiation results from the harsh reactor environment and interactions with the rest of the bundle structure and the reactor pressure tube. Subsequent cooling of the bundle, upon removal from the reactor, results in residual stresses which are reflected in the distortion of the bundle geometry. For CANDU fuel, unlike LWR fuel, the assessment of residual stresses, and therefore subsequent creep strain, must include the effect of interactions between the fuel element and the bundle end-plates via the end-cap to end-plate welds.

In terms of a conventional strain-to-failure criterion for a fuel element, creep is not likely to be a limiting factor for dry storage over a period of 100 years. However, the residual stresses resulting from creep strain on the end plate welding during reactor irradiation and subsequent cooling may present a risk to bundle integrity over long term storage.

5.2 UO_2 Oxidation

Potential oxidation of UO_2 to U_3O_8 in the fuel pellet was considered a concern because the consequent swelling of the pellet would increase stresses on the fuel cladding and may result in cladding deformation or failure.

A model which describes the fundamental data and equations for a detailed model of UO_2 oxidation and provides suggestions for refining and calibrating the model was developed. This model describes the oxidation kinetics of UO_2 pellet fragments by calculating the simultaneous oxidation of grain boundaries, diffusion-controlled oxidation of grains to U_3O_7/U_4O_{9+x}, and further oxidation to U_3O_8 by a nucleation-and-growth process. According to the model results, conditions for detrimental cladding damage from UO_2 oxidation to U_3O_8 are expected to be difficult to reach under a normal storage environment in a DSC (i.e., a conversion yield of 15% to U_3O_8, which would cause cladding diameter increase of 2 %).

During the CEX-1 experiment some used fuel elements from Pickering-A and Bruce-A were deliberately punctured to simulate fuel defects and stored in dry air at 150°C. The fuel elements were monitored over a period of 15 years and the fuel bundles were examined four times during this period. Oxidation of UO_2 resulted in U_3O_8 generation only in the vicinity of the defects. No changes in fuel element diameter were observed in the vicinity of defects. This experiment was conducted under harsher conditions (constant temperature of 150 °C and presence of air) compared to conditions in DSCs. This further supports the assumption that normally there are no conditions for significant U_3O_8 generation in a DSC.

5.3 Stress Corrosion Cracking of Zircaloy (SCC)

Previous studies have indicated that there are two different types of threat from SCC. One potential threat is cracks in the inner surfaces of cladding or end-cap welds induced by the iodine in the gap of the intact fuel. This is largely prevented by the CANLUB coating in fuel elements (early CANDU fuel did not have Canlub coating). The other threat is that of SCC initiating on the outside surface of a fuel element in cases where the DSC cavity becomes contaminated with SCC agents under some hypothetical accident conditions. This case falls under abnormal storage scenarios. To analyze the risk associated with this scenario, it is necessary to determine the maximum predicted concentration of SCC agents in the DSC cavity due to fuel failure.

The results from the AECL studies provide valuable data on the SCC of CANDU fuel and indicate a path for establishing the risk of fuel failure from SCC. Wood's results indicate a clear dependence of SCC on fast-neutron fluences and on the existence of previous notches or marks on the cladding surface. Since both these parameters can be estimated from fuel statistics using available information from PIE of OPG's fuel, an estimate of the risk of cladding failure from SCC could be largely derived based on existing information.

5.4 Hydrogen Effects on Zircaloy (DHC)

The two major potential effects on the fuel Zircaloy components from hydrogen incorporated during irradiation in the reactor are delayed hydride cracking (DHC) and embrittlement, both of which would increase susceptibility to fuel failure when subjected to impact loads or vibration. These phenomena occur as a result of hydrogen and deuterium precipitation in the Zircaloy and they may constitute the biggest concern for long-term integrity of the fuel. The hydrogen and deuterium precipitate in the form of hydrides. Typical hydrogen/deuterium content is in CANDU fuel cladding is below 100 ppm. The morphology and geometric orientation of the hydrides, as well as their size, have a significant effect on the properties of the material.

Temperature cycling of the fuel in storage induced by ambient temperature oscillations may result in a cycle of dissolution and precipitation of the hydrogen in the fuel Zircaloy components. The slow progressive decrease in fuel temperatures during dry storage will eventually result in precipitation of all dissolved hydrogen in the form of hydrides. Coupled with residual stresses in the cladding and welds, and with the temperature gradients within the DSC, these processes could result in migration of the hydrogen to high stress points and in growth or re-orientation of the hydrides (see EPRI 2002, Part II, and Section 4.3).

A report by Rashid et al. (EPRI 2001) presents an analysis defining five conditions that are needed for DHC to be an active mechanism in LWR fuel during dry storage. Based on this study it was concluded that for CANDU fuel, the stress intensity factor is the key parameter to be evaluated in order to assess the potential effect of DHC on used fuel integrity.

6.0 TEMPERATURE MEASUREMENTS AND THERMAL ANALYSES OF USED FUEL IN THE DSC

In 1998, one DSC was instrumented with 24 thermocouples at the inner and outer steel liners and temperature was measured over a period of 100 days. The temperature measurement results were used in 2003/2004 for thermal analysis of used fuel in the DSC. Thermal analysis has included scenarios that are a part of the dry storage processes. Thermal analysis was done for various heat sources, under vacuum and different pressures of helium and air.

In the benchmarking part of the thermal analyses, results were compared with the measured temperatures. The results indicated a difference between the computed and measured temperatures. Differences were on the conservative side because the computed temperatures were higher than measured by approximately 10-15 ^{o}C.

Thermal modeling of the long term dry storage scenarios has shown that the highest temperature of fuel cladding is approximately 140 ^{o}C, during the process of vacuum drying (ANSYS 2004). The seasonal temperature variations of the hottest fuel element were found to be up to 55 ^{o}C. The temperatures were in general lower than the originally predicted and used for evaluation of the used fuel deterioration mechanisms.

7.0 USED FUEL INTEGRITY PROJECT

In 2004, a new five-year plan for investigation of used fuel integrity during long-term (100-year) dry storage was started.

The logic of the approach for the next phase of the used fuel integrity investigation is shown on the block diagram in Figure 1. The plan is to evaluate the characteristics of fuel at the beginning of dry storage based on historic data, and in parallel to investigate envelope of stresses and loads that fuel would undergo after the dry storage period. The results of these two investigations would also be the development of a laboratory examination program for fuel characteristics and for behavior under estimated stresses and loads the fuel would undergo during handling and transport. Fuel stored in ERB, CEX-1 and CEX-2 could be used for some of the examinations. The plans for further used fuel integrity investigations also include additional temperature measurements and thermal analysis.

The results of the program should establish whether used fuel of given characteristics is fit to undergo a certain operation (e.g. transportation). Fuel bundle mechanical integrity is the major concern as it is believed that an occasional pinhole breach of the fuel element is not going to be a major concern during handling and repackaging of the fuel 100 years from now.

The aging processes that will be considered in the Used Fuel Integrity project are those previously noted: cladding creep (including its effect on endplate welds), UO_2 oxidation, SCC and hydrogen effects on Zircaloy-4, as well as mechanical stresses and potential synergistic effects from combinations of these factors. The critical areas of investigation are:

- susceptibility to shock and fatigue failure of Zircaloy components caused by hydrogen mobility and precipitation or re-orientation of hydrides,
- susceptibility to delayed hydride cracking caused by the combined effects of hydrides, residual stresses in the welds and incipient cracks.

Finally, the plan also includes studies of what information that is pertinent to used fuel integrity would have to be preserved for future generations for safe handling and transport of used fuel. The study will also include investigation of the best form to preserve the information.

The specific plan is outlined in more detail below.

7.1 Used Fuel Characteristics at the Beginning of Dry Storage

A project to compile and review all the historic data about the Reference Characteristics of the Used Nuclear Fuel at the beginning of dry storage was started in 2004. The objective of this study is to provide data about fuel characteristics that are pertinent to used fuel integrity, as well as to provide basis for developing further used fuel examination programs.

Preliminary review of data involved categorization of defects that were determined during used fuel inspections. Defects were categorized based on expert experience into moderate and significant risk categories pertinent to losing integrity of the bundle.

Inspection observations were related to the following parameters:

- Residual stress magnitude on the fuel bundle end plate,
- Residual stress magnitude on the weld between the end cap and the end plate,
- Residual stress magnitude on the weld between the end cap and the sheath,
- Residual stress magnitude on the fuel element sheath.

Preliminary data are shown in the Table 2.

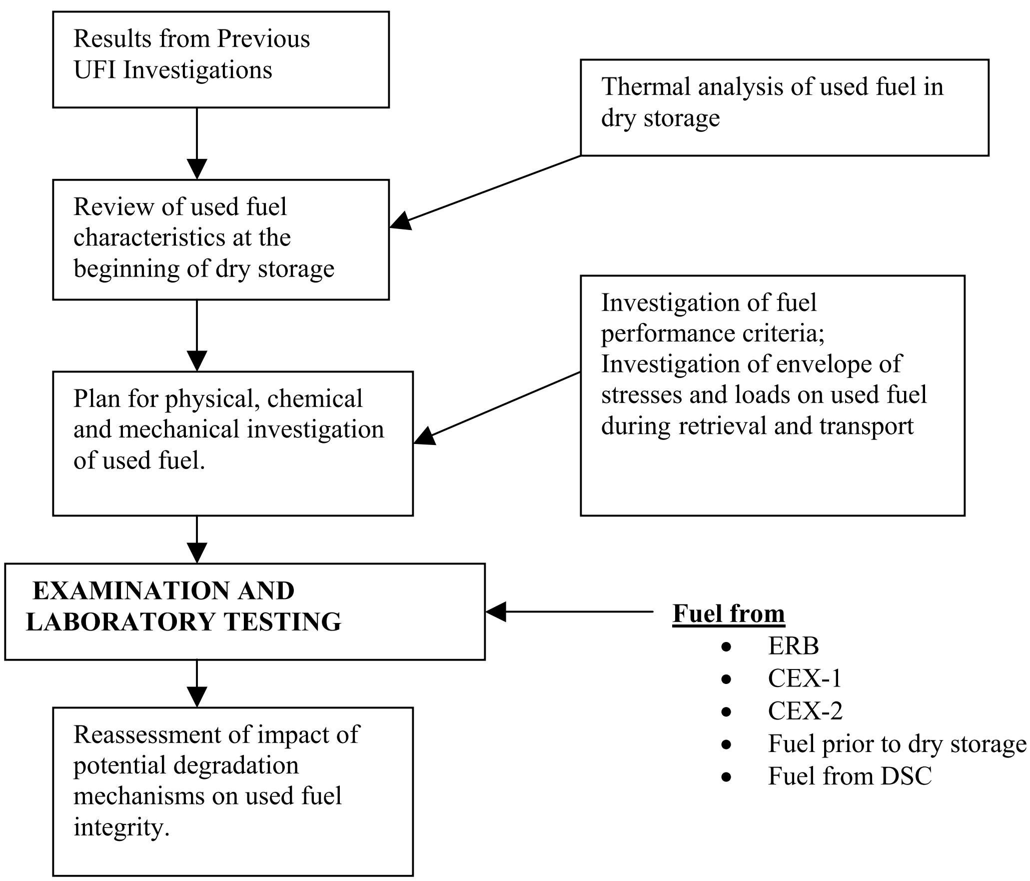

Figure 1: Logic of the approach of the 5-year plan for used fuel integrity investigation

Table 2: Preliminary Data from Fuel Characteristics Evaluation

Total number of used fuel bundles	1,500,000
Number of bundles inspected	9,000
Number of bundles undergone PIE*	~ 300
Estimated % of all bundles with potential loss of integrity	0.02-3.6 (%)

* PIE= Post Irradiation Examination

The wide range of percentage bundles with potential loss of integrity is due to uncertainty in how representative the inspected fuel bundles are of the general population. The range covers from random selection of fuel bundles to targeted inspection based on some incident indications.

7.2 Expected Stresses and Loads during Handling and Transportation

A need to determine the envelope of possible loads and vibrations used fuel may be exposed to during handling and transport after long term storage was identified, and a project to define this envelope was started at the end of 2004. The work will involve review of the current practices and procedures for handling and transport of used fuel. The results of this investigation would enable determination of the acceptable physical characteristics of used fuel that would ensure their integrity during handling and transport.

7.3 Development of a Plan for Examination and Testing of Used Fuel

The examination programs will consider tests and analyses of used fuel that could provide information on stressors and parameters affecting deterioration mechanisms of used fuel. The program will be based on previous examinations and on the identified data gaps for used fuel deterioration modeling. Furthermore, the program will consider testing and examination of used fuel behavior under the envelope of defined loads and vibrations.

8.0 SUMMARY AND PATH FORWARD

Based on the review of technical information on phenomena relevant to fuel integrity and previous investigation and research of CANDU fuel, there are some major conclusions that could be made:

- Under normal storage conditions it is unlikely that CANDU fuel will suffer significant degradation during a period of about 100 years of dry storage. Therefore it should be possible to retrieve, re-package and transport used fuel as required using methods and systems similar to those used today.
- To provide increased confidence regarding the first conclusion, additional investigation will be conducted in areas where there is a higher uncertainty in the prediction of fuel condition and on some degradation processes to which the fuel appears to be more vulnerable.
- Additional work will also be conducted to evaluate the possible effects of abnormal and/or accidental conditions on the integrity of fuel.

9.0 REFERENCES

ANSYS. 2004. Thermal Modeling of OPG Used Fuel Dry Storage Container; Report 4 - Long-Term Storage Scenarios. OPG Report 00104-REP-02308-00004 R00.

EPRI. 2001. Fracture Toughness for Zirconium Alloys: Application to Spent Fuel Cladding in Dry Storage. EPRI Report 1001281.

EPRI. 2002. Dry Cask Storage Characterization Project-Final Report. EPRI Report 1002882.

Frost C. R. 1994. Current Interim Used Fuel Storage Practice in Canada. Ontario Hydro Report N-03710-940052.

McMurry J., Dixon D.A., Garroni J.D., Ikeda B.M., Stroes-Gascoyne S., Baumgartner P. and Melnyk T. W. 2003. Evolution of a Canadian Deep Geologic Repository: Base Scenario. OPG Report 06819-REP-01200-10092-R00.

Wasywich, K.M. and Frost C.R. 1991. Examination of Used CANDU Fuel Following 27 years of Storage under Water. Proceedings 3rd International Conference on Nuclear Fuel Reprocessing and Waste Management (RECOD 91), Sendai, Japan.