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BHABHA ATOMIC RESEARCH CENTRE
MUMBAI, INDIA

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21	Affiliation of author (s) :	Reactor Engineering Division, Bhabha Atomic Research Centre, Mumbai
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60 Abstract : One of the strategies employed for management of spent fuel prior to their final disposal/reprocessing is their dry storage in casks, after they have been sufficiently cooled in spent fuel pools. In this interim storage, one of the main consideration is that the fuel should retain its integrity to ensure (a) radiological health hazard remains minimal and (b) the fuel is retrievable for down stream fuel management processes such as geological disposal or reprocessing. For dry storage of spent fuel in air, oxidation of the exposed UO₂ is the most severe of phenomena affecting the integrity of fuel. This is kept within acceptable limits for desired storage time by limiting the fuel temperature in the storage cask. The limit on the fuel temperature is met by having suitable limits on maximum burn-up of fuel, minimum cooling period in storage pool and optimum arrangement of fuel bundles in the storage cask from heat removal considerations. The oxidation of UO₂ by moist air has more deleterious effects on the integrity of fuel than that by dry air. The removal of moisture from the storage cask is therefore a very important aspect in dry storage practice. The kinetics of the oxidation phenomena at temperatures expected during dry storage in air is very slow and therefore the majority of the existing data is based on extrapolation of data obtained at higher fuel temperatures. This and the complex effects of factors like fission products in fuel, radiolysis of storage medium etc. has necessitated in having a conservative limiting criteria. The data generated by various experimental programmes and results from the on going programmes have shown that dry storage is a safe and economical practice

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1.0 INTRODUCTION

The management of nuclear fuel waste is an important aspect in the nuclear power programme. Spent fuel removed from reactor itself is treated as waste by many countries, but in the context of our nuclear power programme, spent fuel is reprocessed to separate out useful materials (Uranium and Plutonium) and the remaining radioactive material is treated as the waste. The disposal of nuclear fuel waste is done by long term waste management procedures.

The basic characteristics of spent fuel is that it is radio-active and it generates heat due to absorption of considerable fraction of radiation energy in the fuel. The radioactivity in the spent fuel decreases with time and with it the generation of heat in fuel also decreases. The spent fuel bundles are therefore stored for few years depending on the burnup of fuel and waste management programme until the radioactivity and heat generation drops to levels suitable for reprocessing or permanent disposal.

The spent fuel bundles are stored under water in storage pools located adjacent to the reactor. The water in the storage pool provides radiation shielding and removes heat from the fuel bundles. This water in turn is cooled by heat exchangers and purified to remove any contaminants. The pools are monitored for degradation and leaks.

The storage pools for PHWRs have been designed for a capacity to store bundles expected to be removed in ten years of operation. For additional requirement in the storage capacity, dry storage of spent fuel at reactor sites is more economical than augmentation of wet storage capacity by construction of additional storage pools. After the spent fuel bundles have been sufficiently cooled in storage pools, they can be transferred into concrete storage casks. The radiation is shielded by concrete and the heat is transferred through the storage medium and concrete wall to outside air. The storage medium inside the cask can be either inert gas (helium, argon) or any other gas like air, nitrogen, carbon-dioxide.

This note is based on available information in the published literature and gives an overall picture of aspects related to fuel in dry storage. It describes the major concerns during dry storage, fuel integrity criteria and review of dry storage technologies of LWR and CANDU fuels.

2.0 MAJOR CONSIDERATIONS FOR ADOPTION OF A DRY STORAGE SCHEME

The primary safety concerns which have to be satisfactorily addressed for dry storage of spent fuel are :

- (a) Adequate fuel cooling.
- (b) Effective containment of radioactive release in the event of fuel failure.
- (c) Adequate radiation shielding.
- (d) Adequate physical security
- (e) Long term structural integrity.

Among the mentioned issues, one of the main considerations pertaining to fuel is that the integrity of fuel should be retained to ensure (a) radiological health hazard remains minimal and (b) fuel is retrievable for down stream fuel management processes such as geological disposal or reprocessing. The major problem being faced in the evolution of suitable criteria to ensure fuel integrity is that long term experimental programmes spanning several decades are required to generate data for conditions prevailing during dry storage. This has essentially resulted in availability of very little data in quantitative terms for factors which have an influence on fuel integrity and therefore the limiting criteria used are very conservative.

3.0 FUEL INTEGRITY CRITERIA DURING DRY STORAGE

The loss of fuel integrity during dry storage basically means damage of the clad to an extent which would affect the retrieval and subsequent handling of fuel or pose radiological problems. Fuel integrity criteria have been evolved on the basis of various phenomena affecting the integrity of fuel. The storage medium in the cask has a strong influence on the phenomena affecting the integrity of fuel. Hence, fuel integrity criteria are different for different storage mediums.

3.1 Phenomena Affecting the Integrity of Fuel

The damage to zircaloy clad of UO_2 fuel can occur due to any of the following :

- (a) Clad creep under internal gas pressure.
- (b) Stress Corrosion Cracking (SCC) or Delayed Hydride Cracking (DHC) of clad.
- (c) Corrosion of clad due to oxidation.
- (d) Defect propagation of clad due to oxidation of UO_2 , in case of a defective fuel.

3.2 Influence of Storage Medium

There has been mainly two approaches as regards to the storage medium in dry storage of UO_2 fuels. In one of the approaches, which has been especially used for high burnup LWR fuels, Helium is the storage medium and in the other which has been used for low burnup CANDU fuel, air is the storage medium. The essential difference between them is that the former allows higher fuel temperatures during dry storage, but the latter is a simpler dry storage practice. The higher allowable temperature is necessary for LWR fuels due to their higher decay heat generation. However, storage of CANDU fuel in Helium has also been done to take advantage of its better heat transfer characteristics and thereby make the storage arrangement more compact. Appendix-I gives a comparison of the heat transfer characteristics of a few storage mediums.

3.3 Basis for Fuel Integrity Criteria with Helium as Storage Medium

Helium due to its inert nature does not cause oxidation of UO_2/Zr -clad. The other phenomena (clad creep and SCC/DHC) have been used as the basis for evolving the fuel integrity criteria.

3.4 Basis for Fuel Integrity Criteria with Air as Storage Medium

The phenomenon of oxidation of UO_2 has been used as the basis for evolving the fuel integrity criteria. This is because oxidation of UO_2 fuel is the most severe of the phenomena affecting the integrity of fuel in this case. SCC/ DHC and clad creep are not expected to pose much problem, once the criteria to prevent loss of fuel integrity due to oxidation of UO_2 is met[1]. In addition, the defective fuel has been taken as the basis for forming the regulatory criteria due to the following reasons:

- (a) Damage to clad by propagation of defect due to oxidation of UO_2 has been found to be more severe than direct Zr -clad oxidation.
- (b) There is possibility of defective fuel coming out of the reactor without being detected by the failed fuel detection system.
- (c) Damage to clad can take place at any stage during handling or storage in the pool.

4.0 REVIEW OF FUEL INTEGRITY CRITERIA USED FOR VARIOUS FUELS

The limiting criteria for dry storage of spent fuel can be either empirical or mechanistic. In empirical form, the limiting criteria are expressed in the form of maximum allowable fuel temperature, storage medium and duration of dry storage. Table-1 gives this form of limits employed by various regulatory agencies [2,3]. In the mechanistic form, the limiting criteria are directly in terms of the phenomena affecting the integrity of fuel i.e. for storage of UO_2 fuel in dry air, the criterion can be directly a limit on the maximum allowable oxidation. Table-2 shows the limits in the mechanistic form and the basis of this criterion is discussed in Section 5.0 [4].

5.0 OXIDATION OF UO_2 IN AIR

A thermodynamic assessment of the compatibility of UO_2 with air shows that chemical reactions of UO_2 is possible only with oxygen and not with pure nitrogen. UO_2 when heated in air readily oxidises and initially forms U_4O_9/U_3O_7 before eventually forming low density phase U_3O_8 along with powdering of the oxide. The state of the oxide can be identified by the crystal structure through X-ray diffraction or the U_6^+/U_4^+ ratio measured by X-ray Photo electron Spectroscopy.

Table-3 gives the densities of various oxides of uranium. A 100% conversion of UO_2 to U_3O_8 leads to about 36% increase in volume. The oxidation of fuel can therefore cause swelling and splitting of fuel clad which could complicate the subsequent handling of the bundle. Experimental observations for CANDU fuel indicate that there is no splitting of clad for clad diameter increase below 2% (a 15% conversion of UO_2 to U_3O_8 can lead to this 2% clad dia. increase). Based on this, a very conservative criterion for maximum acceptable degree of oxidation can be 1.5% conversion of an exposed fuel surface. The advantage of this form of criterion is that it is experimentally verifiable (Table-2 gives the time-temperature relationship for this form of criterion based on the assumption that only U_3O_8 is formed during the oxidation of UO_2) [4].

TABLE -1

CRITERIA EMPLOYED BY VARIOUS REGULATORY AGENCIES FOR DRY STORAGE OF SPENT FUEL

TYPE OF FUEL	COUNTRY	STORAGE ENVIRONMENT	LICENSED TEMP. (° C)
LWR FUEL	USA	HELIUM	380
	GERMANY	HELIUM	410
CANDU FUEL	CANADA	DRY AIR	160
		HELIUM	250
MAGNOX FUEL	UK	DRY AIR	150
	ITALY	CO ₂	300
		NITROGEN	Not Available.

TABLE -2

ESTIMATED TIMES FOR 1.5% SURFACE CONVERSION OF UO₂ TO U₃O₈ *

T (°C)	LOWER ESTIMATE (90% confidence)		UPPER ESTIMATE (90% confidence)	
	t(hrs)	t(yrs)	t(hrs)	t(yrs)
	100	7.2 x 10 ⁶	820	8.8 x 10 ⁷
150	3.4 x 10 ⁴	3.8	2.7 x 10 ⁵	31
170	5.4 x 10 ³	0.6	3.9 x 10 ⁴	4.5
200	460		3000	
250	13		83	
300	0.6		5.3	

* Based on the regression analysis illustrated in Fig.-2

TABLE- 3

DENSITIES OF VARIOUS OXIDES OF URANIUM

	UO ₂	U ₃ O ₇	U ₃ O ₈	UO ₃ .x.H ₂ O
DENSITY(g/cc)	10.97	11.4	8.35	
Volume Change for 100% Conversion	---	2% decrease	36% increase	> 160% increase

The rate of oxidation is basically a function of temperature of fuel, but the oxidation mechanism is influenced by factors like presence of fission products in fuel, moisture in air and products formed due to radiolysis. The effect of the above mentioned factors is quite complex and therefore for a correct assessment of oxidation phenomena, the experiment has to preferably simulate all the factors simultaneously.

5.1 Effect Of Temperature[4,5]

The rate of oxidation increases with temperature. Time-Temperature relation for 2% increase in clad diameter due to oxidation of UO_2 to U_3O_8 is given in Fig.-1. At temperatures prevalent during dry storage, for every 25-30°C rise of UO_2 temperature, the rate of oxidation increases by about 10 times. As the oxidation rates are very slow at temperatures below 200°C, the data generation of oxidation rates at temperatures below 200°C relies on extrapolation of kinetic data from elevated temperatures (a plot of the rate constant for formation of U_3O_8 is given in Fig.-2).

5.2 Effect of Moisture [6,7,8]

The moisture in air affects the oxidation of UO_2 in essentially two ways (i) as a reagent for the formation of hydrated uranium oxides ($\text{UO}_3 \cdot x\text{H}_2\text{O}$) and (ii) as a potential source of radiolytic oxidants of H_2O . The density of the hydrated uranium oxides are very low and therefore conversion of UO_2 to $\text{UO}_3 \cdot x\text{H}_2\text{O}$ can cause unacceptable swelling and splitting of the fuel clad (A 100% conversion of UO_2 to $\text{UO}_3 \cdot x\text{H}_2\text{O}$ can lead to about 160% increase in volume). The formation of the hydrated uranium oxides is essentially by moist oxygen. The thermodynamics of chemical reactions indicate that moist nitrogen is in equilibrium with fuel stoichiometry of $\text{UO}_2 \cdot 0.06$ and therefore the very low density phases of oxides of uranium will not be formed by oxidation of UO_2 by moist nitrogen. The effect of moisture due to its radiolysis is discussed in Section 5.3.

In addition, experimental results indicate that the mechanism of oxidation for moist air is different from that for dry air. In dry air, oxidation is localised mainly in the defect region. It occurs on exposed UO_2 surfaces and penetrates into the grain of UO_2 matrix. In contrast, oxidation in moist air is distributed through out the length of the fuel and it proceeds along the grain boundaries before penetrating into the grain.

5.3 Effect of Radiation [8,9]

The gamma field due to radio-activity of spent fuel has an effect on the oxidising potential of the storage medium. Radiolysis of nitrogen in the presence of oxygen or moisture generates N_xO_y compounds and moisture itself becomes a source of O_2 and H_2O_2 in a radiation field. The thermodynamics of chemical reactions indicate that the compounds- oxides of nitrogen and H_2O_2 will oxidise UO_2 to low density oxide phases. Some of the experimental studies carried out to study the effect of radiation are discussed in Section 6.0. (Most experiments carried out on small samples of irradiated fuel specimens in laboratories underestimate the oxidation rates due to the absence of free radicals/compounds produced due to radiolysis).

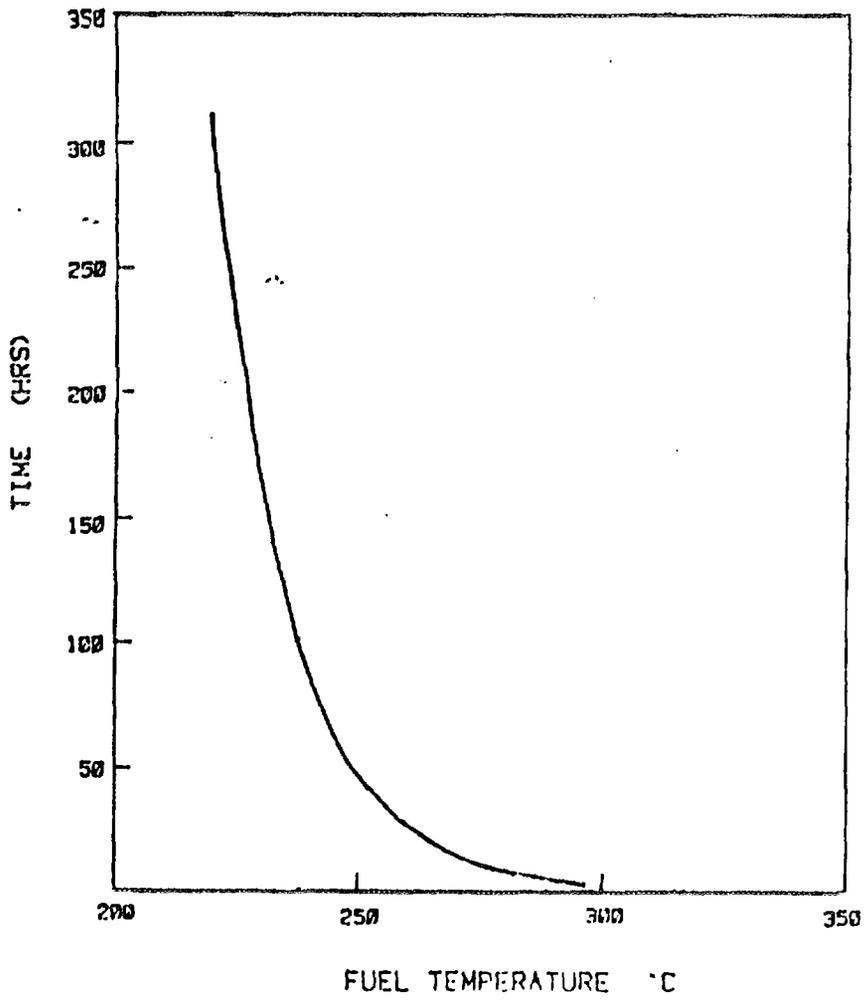


FIG. 1

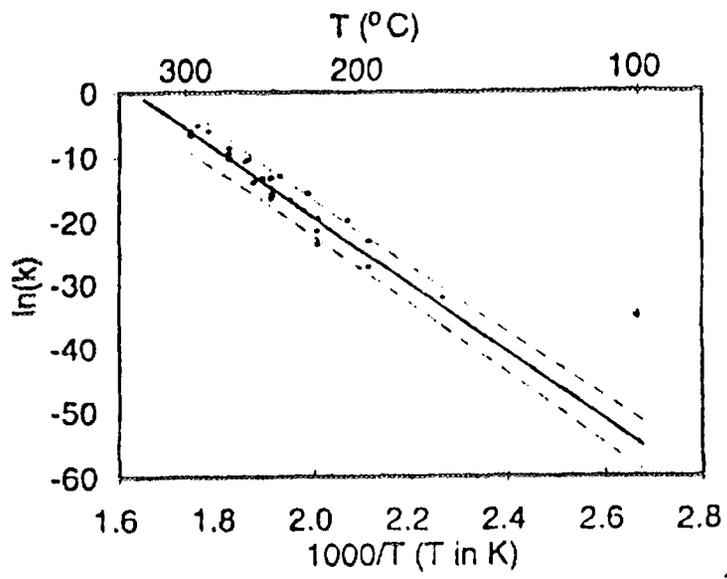


FIG. 2

5.4 Effect of Fission Products [4,10] :

Experiments have indicated that there is a gradual suppression of U_3O_8 formation with increasing burnup for e.g., defected CANDU fuel elements (1 atom % burnup) have been reported to suffer major swelling and splitting after about 200 hrs at 250°C, whereas irradiated LWR fuel fragments (2-5 atom % burnup) showed no sign of U_3O_8 formation after 10,000 hrs at the same temperature. The effect of increase in the fraction of fission products (burnup) on U_3O_8 formation can be seen in Fig.-3. Fission products seem to also have an effect on the mechanism of oxidation. In un-irradiated UO_2 , air-oxidation proceeds from the surface inwards by the formation of a thin layer of U_4O_9/U_3O_7 , while spent fuel oxidise internally by rapid advance of U_4O_9/U_3O_7 along grain boundaries and then penetrate into the UO_2 grains.

6.0 REVIEW OF TECHNOLOGIES FOR DRY STORAGE

6.1 CANDU FUEL [1,4,9]

AECL initiated an experimental program in 1978 to investigate the long term behaviour of used fuel (both undefected and defected) during dry storage. This program which is scheduled to continue beyond the year 2000 basically consists of (i) Easily Retrievable Basket (ERB) experiment (dry storage in air at seasonally varying temperatures) and the Controlled Environment Experiments, CEX-1 (storage in 15°C dew point dry air at 150°C) and CEX-2 (storage in moisture saturated air at 150°C). The fuel bundles are monitored and examined at regular intervals during the experimental programme.

6.1.1 ERB Experiment

The ERB experiment involves the storage of two typical undefected used fuel bundles from Ontario Hydro's Pickering Nuclear Generating Station. It was loaded in 1978 after pre-storage characterisation of the bundles. The temperature of the bundles have been recorded and documented since loading. The fuel temperature have dropped even to temperatures below 0°C (upto -19°C) during winters. The air in the ERB is monitored for radioactive material to detect any defect of the bundles and no radioactive release has been detected till now.

6.1.2 CEX Experiments

CEX-1

The CEX-1 experiment involves storage of eight bundles, four from Pickering and four from Bruce. The outer elements in two of Pickering and two of Bruce bundles were intentionally defected before storage by drilling a 3 mm. dia. hole to compare the relative storage behaviour of undefected and defected elements. The temperature of dry air in the CEX-1 experiment is more than in the ERB experiment. The bundles are stored in a carbon steel container called "basket". The basket has two isolated compartments, one containing the four intact bundles and the other containing the four intentionally defected bundles. Each bundle is held inside an open ended carbon steel container within the basket. The basket is then stored in an insulated, electrically

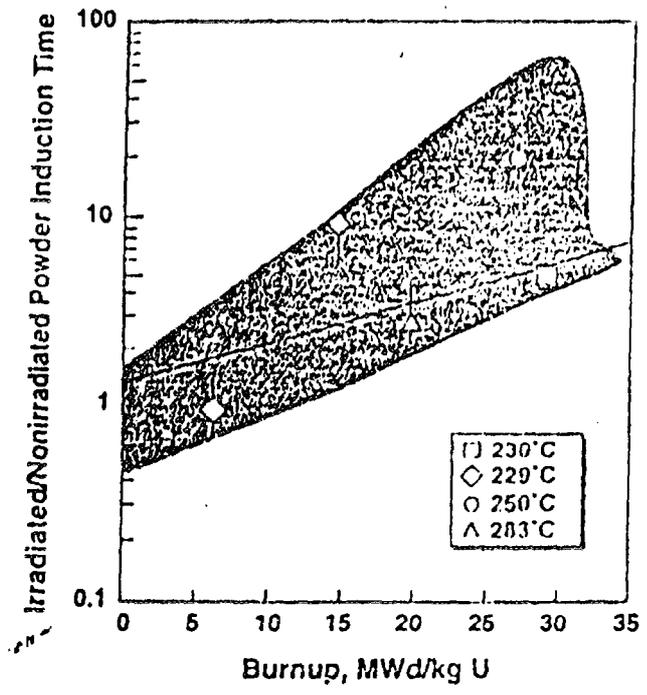


FIG. 3

heated chamber in a concrete canister. The free void volume of the defected bundle compartment is 42.1 litres.

CEX-2

The CEX-2 experiment also involves storage of eight bundles, four from Pickering and four from Bruce. The outer elements in two of Pickering and two of Bruce bundles were intentionally defected before storage by drilling a 3 mm. dia. hole to compare the relative storage behaviour of undefected and defected elements. Moist environment is being used in CEX-2 to simulate a situation in which water might be transferred from water pools to the container in which the bundles are to be dry stored. Each fuel bundle is stored in an individual Type 304 stainless steel pressure vessel with 100ml of distilled, de ionised water to saturate the air with moisture. The pressure vessels are stored in a basket and a concrete canister similar in design as used in CEX-1. The free void volume of the pressure vessel is 4.7 litres.

Results From CEX Experiments

The intermediate examinations of fuel bundles indicate that oxidation in CEX-2 was more pervasive along the grain boundaries than in CEX-1, but was limited to a thin surface film on the grains. Oxidation in CEX-1 was more limited in its progress along an element from the defect, but produced thick layers (>1 micron) of U_3O_7/U_4O_9 on UO_2 grains in the defect region. No U_3O_8 was detected in either experiment after 100 months of operation with CEX-1 and 69 months with CEX-2.

The analysis of the container atmosphere in the CEX experiments showed significant oxygen depletion (Table-4 gives the results of the analysis after a storage period of 58.5 months in CEX-1 and 39.5 months in CEX-2. The results could have been affected by ingress of outside air into the CEX-1 defected bundle compartment due to a leaking swagelok fitting). The inferences that can be drawn from the results of the analysis are:

- (i) Least depletion of oxygen took place in the intact bundle compartment of CEX-1 and the high concentration of CO_2 indicates that a considerable amount of oxygen was consumed by the container components.
- (ii) Greater depletion of oxygen occurred in the defected bundle compartment of CEX-1, whereas the concentration of CO_2 increased only slightly which indicates that O_2 was mainly consumed by fuel oxidation.
- (iii) The increase in concentration of hydrogen in CEX-2 indicates radiolysis of moisture and it has contributed around 1/3 of the total oxygen consumed by the container components and UO_2 fuel.
- (iv) All oxygen was completely consumed in the CEX-2 compartment. The increase in CO_2 concentration and total depletion of oxygen indicates significant consumption of oxygen by both fuel (about 75% of oxygen) and the container components (about 25% of oxygen).

Due to the large number of defected elements, the supply of oxygen relative to the quantity of UO_2 was less in the CEX experiments than that expected in a commercial scale fuel storage basket. This is because the fuel storage basket is unlikely to contain more than one defected

TABLE - 4

**ANALYSES OF THE ATMOSPHERES FROM THE STORAGE VESSELS
CONTAINING CEX-1 AND CEX-2 FUEL BUNDLES**

COMPONENT	STORAGE ATMOSPHERE (VOL %)			NORMAL AIR (VOL %)
	* CEX-1		** CEX-2	
	(a)	(b)		
H2	<0.01	<0.01	3.90	<0.02
HELIUM	0.14	<0.01	0.13	<0.02
N2	92.3	79.2	88.8	78.1
O2	6.14	12.4	<0.02	20.9
ARGON	1.09	2.03	1.11	0.93
CO2	0.35	6.33	5.19	0.03
KRYPTON	<0.05	<0.05	0.18	<0.02
XENON	<0.05	<0.05	0.68	<0.02

In this table ,(a) and (b) refer to defected and undefected bundle compartment respectively.

* The duration of the CEX-1 second stage period was 58.5 months in dry air at 150°C whereas the total storage time to the end of second storage period was 99.5 months.

** The duration of the CEX-2 second storage period was 39.5 months in moisture-saturated air at 150°C, whereas the total storage time to the end of the second storage period was 69 months.

element and therefore sufficient oxygen would be available to oxidise the exposed UO_2 entirely to U_3O_8 . To help simulate commercial fuel storage conditions more accurately, the CEX-1 experiment was modified in 1989 to increase the volume of air available to fuel. The available volume of air was thereby increased from 0.8 litres to 48 litres per defected element. The examination of fuel elements after 40 months of exposure to the increased air supply (following the 100 months in limited air) at 150°C has revealed traces of U_3O_8 in the defected fuel elements.

In 1992, a new experiment (Alternate Controlled Experiment, ACX) was initiated, in which selected elements from the CEX-2 fuel have been exposed to moisture-saturated air at 150°C with replenishment of air supply at intervals of about 20 days. The results of intermediate examinations of the ACX experiment are not available.

6.1.3 Laboratory Tests

Oxidation of CANDU nuclear fuel (UO_2) heated at 150°C in air, in O_2 with 60% saturated steam and Ar with 60% saturated steam atmospheres for 2 years in gamma fields of dose rate ~ 1500 R/hr was investigated using X-ray photoelectron spectroscopy, X-ray diffraction and scanning electron microscopy. Surface analysis of the UO_2 disks exposed to air showed formation of U_3O_8 , though the rate of U_3O_8 formation was very low. The presence of water vapour along with O_2 increased the oxidation of UO_2 in gamma fields, leading to the formation of U_6^+ phases i.e. $\text{UO}_3 \cdot x\text{H}_2\text{O}$ along with U_3O_8 . This is due to the presence of oxidants produced by the radiolysis of water. However UO_2 did not suffer any oxidation by water vapour radiolysis at 150°C in an O_2 free atmosphere. This is different from various other observations at room temperatures, where the radiolysis of water has been shown to cause UO_2 oxidation even in the absence of O_2 . This deviation has been attributed to the increased reactivity of the reductants (also produced by radiolysis of water) at higher temperatures, which has cancelled the oxidation potential of the oxidants. Table-5 gives the XPS and XRD results of the experiment.

6.2 LWR FUEL [8]

An assessment of nitrogen as an atmosphere for dry storage of spent fuel was done at Pacific Northwest Laboratory (PNL). This was to demonstrate the suitability of nitrogen as a substitute for Helium. Tests were carried out on bare UO_2 fuel pellets under conditions expected in LWR dry storage practice with nitrogen environment. The nitrogen environment contained low concentrations of impurities such as moisture, oxygen and radiolytic products, as the inventories of these impurities are expected to be kept low in the hermetically sealed casks by proper dry storage practice. The tests were carried out under the following conditions:

Non-irradiated pellets in Air+1 vol% NO_2

This was done to study the oxidation behaviour of radiolytic products of the air/nitrogen. The test showed that oxidation at 250°C in air+1 vol% NO_2 was considerably more than in air at the same temperature.

TABLE - 5

SURFACE OXIDATION OF UO₂ DISKS AT 150°C IN GAMMA FIELD
XPS and XRD RESULTS

Experiment Type	Atmosphere ^a	Sample	XPS Results U ⁶⁺ /U ⁴⁺	XRD results: oxide phases seen ^d
A	Air, open	M	2.8	UO ₂ , U ₃ O ₇ , U ₃ O ₈ , U ₈ O ₁₉
		N	3.7	UO ₂ , U ₃ O ₇ , U ₃ O ₈ , U ₈ O ₁₉
		S	2.6	UO ₂ , U ₃ O ₇ , U ₃ O ₈ , U ₈ O ₁₉
B	Air, closed	P	5.6	UO ₂ , U ₃ O ₇ , U ₃ O ₈ , U ₈ O ₁₉
		Q	10	UO ₂ , U ₃ O ₇ , U ₃ O ₈ , U ₈ O ₁₉
		L	6.2	UO ₂ , U ₃ O ₇ , U ₃ O ₈ , U ₈ O ₁₉
C	O ₂ + 60%ss	W	11	UO ₂ , U ₃ O ₇ , U ₃ O ₈ , UO ₃ , UO ₃ .x.H ₂ O
		X	118	UO ₃ .x.H ₂ O, UO ₃ , U ₃ O ₇ , UO ₂ , U ₃ O ₈
		Z ^b	2.2	U ₃ O ₇ , UO ₂ , U ₃ O ₈ , U ₈ O ₁₉ , UO ₃ (?)
D	Ar + 60% ss	V	0.01	UO ₂
		O ^c	0.30	UO ₂ , U ₃ O ₇ , U ₁₆ O ₃₇

^a ss means saturated steam at 150°C.

^b This experiment was probably equivalent to A.

^c This sample's atmosphere probably contained a trace of air.

^d The phases are listed in order of decreasing intensity in the XRD pattern.

Non-irradiated pellets in nitrogen

Bare non-irradiated pellets were placed in three capsules that were evacuated and backfilled with nitrogen (20°C dew point) at 225°C, 250°C and 275°C in a radiation field of 200,000 R/hr. The gas volume in each container was 163 cu.cm. and the oxygen concentration was less than 0.01 vol%. The moisture present in 20°C dew point nitrogen is 0.017mg/cm³ which corresponds to the presence of 2.8 mgs. of moisture in the capsule. No measurable changes in weight or appearance of UO₂ pellets was observed after 1165 hrs of testing.

Spent fuel fragment tests in nitrogen

Tests were conducted in a radiation field of 12000 R/hr with 95 gms of spent fuel at 275°C fuel temperature, 20°C dew point nitrogen environment and with 170 gms of spent fuel at 380°C fuel temperature, 30°C dew point nitrogen environment. The volume of oven used in the test was 16,000 cu.cm. No significant changes in weight or appearance of the fragments was observed after 7 weeks of testing.

7.0 INTERIM DRY STORAGE PRACTICE FOR CANDU FUELS [3,11,12,13,14]

The spent fuel coming out of the reactor is initially stored in storage racks in water pools (different types of storage racks used are shown in Fig.-4). After the decay heat output of the spent fuel has fallen sufficiently, interim storage is continued in a dry storage facility at some of the reactor sites. Table-6 gives the status of dry storage technology in Canada and Table-7 gives the salient features of their dry storage practice.

7.1 Storage in Concrete Canisters

At Whiteshell Nuclear Research Establishment (WNRE), Concrete Canisters (CC) have been licensed to store fuel to a maximum temperature of 250°C in helium atmosphere and at other concrete canister (CC) facilities (Gentilly-1, Douglas point), the CCs are licensed for the storage of spent fuel to maximum temperature of 160°C in dry air (CC used is shown in Fig.-5). The fuel at WNRE is transferred from water pool to a hot cell facility, where it is dry loaded in vertical position into fuel storage containers called baskets. At the other CC facilities, the storage tray is tilted from horizontal to vertical position (along with the bundles) and using the bundle lifting tool, the bundles are removed from the tray and loaded into the basket in the pool. After the basket has been filled to capacity, the cover is placed over the basket and it is taken to a Shielded Welding Station. Here the fuel bundles and the basket are dried by means of hot air blast and the cover is seal welded to the base plate of the basket. The baskets are then loaded into CC at the Dry Fuel Storage Facility by means of Fuel Transport Flask. After the CC has been filled to capacity with loaded baskets, the canister lid is positioned and seal welded to the CC liner to form the second barrier (double containment).

TABLE- 6

STATUS OF DRY STORAGE TECHNOLOGY IN CANADA

SITE	LOCATION	STORAGE METHOD	INITIAL LOADING	ATMOSPHERE	t U (apx.)	STATUS
Whiteshell	Manitoba	CC	1975	Air or He	24	Continuing
Gentilly	Quebec	CC	1985	Air	67	Continuing
Douglas Point	Ontario	CC	1986	Air	300	Continuing
CRNL	Ontario	CC	1988	Air	65	Continuing
CRNL	Ontario	Tile holes	1960s	Air	85	Continuing
Pickering-A	Ontario	DSC	1988	He	7.6	Unloaded
Pickering-A	Ontario	DSC	1990	He	7.7	Continuing
Point Lepreau	New Brunswick	CC	1991	Air	358	Continuing
Gentilly	Quebec	CANSTOR	1995	Air	--	Planned

TABLE- 7

SALIENT FEATURES OF CANADIAN DRY STORAGE PRACTICE

TYPE OF CONTAINER	BUNDLE ORIENTATION	DRYING PROCESS	STORAGE ATMOSPHERE	LICENSED TEMP
CONCRETE CANISTER	VERTICAL	HOT BLAST OF AIR	DRY AIR	150° C
DRY STORAGE CONTAINER	HORIZONTAL	VACUUM DRYING	HELIUM	250° C

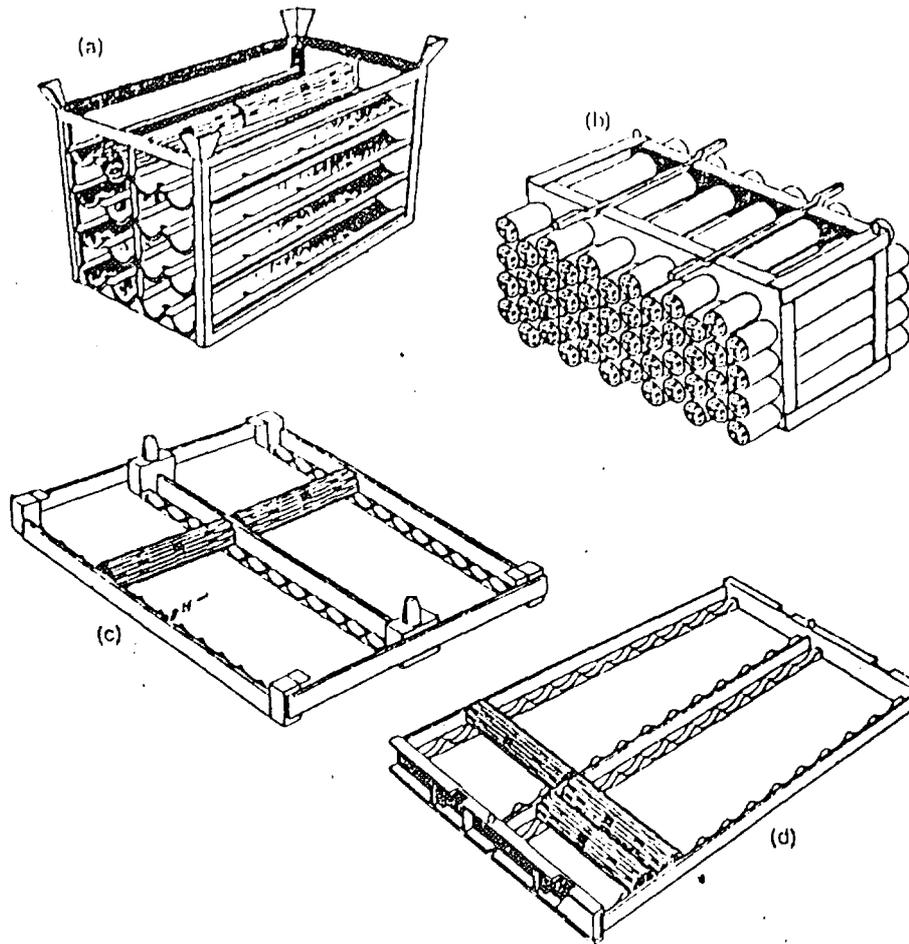


FIG. 4 CANDU spent fuel wet storage containers. (a) The Pickering nuclear generating station spent fuel storage basket (32 bundle capacity); (b) spent fuel shipping and storage module (96 bundle capacity); (c) Bruce nuclear generating station spent fuel storage tray (24 bundle capacity); (d) CANDU 600 MW(e) spent fuel storage tray (24 bundle capacity).

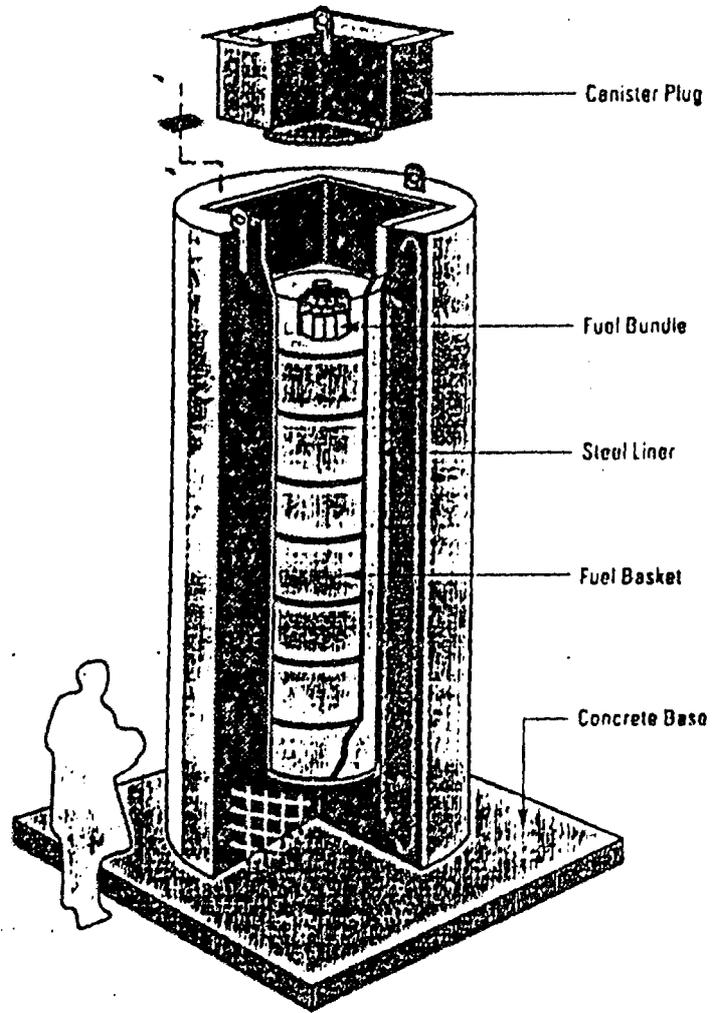


Figure 5a Concrete Canister

The Basket Cover is Seal-Welded to the Base Plate Centre Post to provide Containment

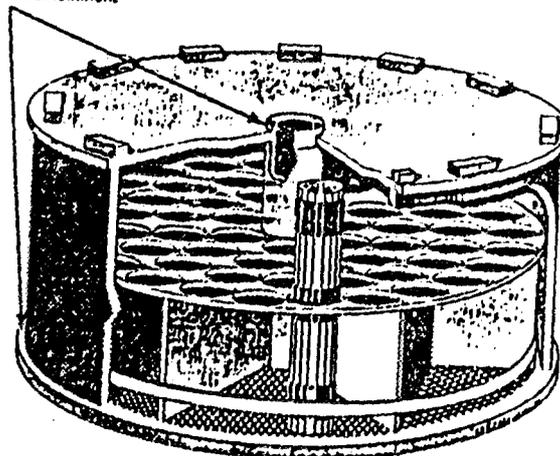


Figure 5b Spent Fuel Storage Basket Containing 60 Bundles

The CC at Douglas point has heat release of less than 1.5 KW although it has a thermal licence of 4.4 KW. Table-8 gives the results of thermal aspects and Table-9 gives the results of radiation shielding aspects of the Douglas Point storage programme.

7.2 Storage in Dry Storage Container

At Ontario Hydro, the fuel bundles are loaded into fuel modules in the water pool. The modules are directly loaded into Dry Storage Container (DSC) under water (DSC shown in Fig.-6). The DSCs are then removed from the pool and water is drained from the cask. It is then vacuum dried, seal welded, backfilled with Helium and transported to the storage site dry tractor trailer.

During the commissioning and initial operation of the Pickering dry storage programme with DSCs, the amount of water left in the container after draining was found to be about 55 litres against the expected 10 litres. Vacuum drying was done basically in two stages. In the first stage vacuum drying was connected and allowed to run for 14 hours. Water was continuously removed and internal pressure was lowered from atmospheric pressure to 13 mbar(a). The DSC was then transported to the storage area. Here about 20 litres of water was collected through the drain port and it is believed that this water was forced out from the module tubes by vibrations during the transport. The second stage of vacuum drying was then carried out for about 4 days and internal pressure of 5.5 mbar(a) was achieved. This was followed by final vacuum drying for about 2 hours with internal pressure reaching 1 mbar(a) before backfilling with helium to 930 mbar(a).

8.0 REVIEW OF MOISTURE REMOVAL ASPECTS

The primary sources of moisture in the storage medium of the cask are due to the following:

- (a) Residual moisture remaining in the storage system after incomplete drying from pool operations.
- (b) Desorption of moisture from crud on the surface of fuel rods and outgassing of storage component surfaces.
- (c) Release from water-logged reactor breached fuel rods.

8.1 Importance of Moisture Removal For Storage in Helium

The licensed fuel temperatures for storage in Helium is as high as 400-420°C. At this temperature the rate of oxidation is very high and complete oxidation of UO_2 can take place within few hours. This requires that oxidation of UO_2 has to be almost completely avoided.

The drying operation in the LWR dry storage practice is therefore tuned to meet this very stringent requirement. The moisture content in the Helium filled in the storage casks is kept to a minimum (dew point of storage medium is about -20°C to -40°C). The results from characterisation tests of their drying practice showed that the residual moisture after vacuum drying in the entire cask due to the above mentioned sources of moisture (a) & (b) is less than

TABLE- 8

MAXIMUM TEMPERATURE IN DOUGLAS POINT CANISTERS

LOCATION	CALCULATED	MEASURED
DAILY AVERAGE AMBIENT AIR	30	30
CANISTER SURFACE	46	32
CANISTER LINER	73	51
BASKET WALL	97	67*
CLAD TEMPERATURE	124	84*

* extrapolated

Calculations based on a windless, hot summer day, having an average ambient temperature over 24 hrs of 30°C.

TABLE- 9

MAXIMUM DOSE RATE AT VARIOUS LOCATIONS

LOCATION	DOSE RATE (mSv/h)
ON CONTACT WITH WELDING STATION	0.05-0.1
ON CONTACT WITH TRANSFER FLASK	0.1 - 0.22
ON CONTACT WITH ONE CANISTER	0.025 - 0.064
BETWEEN 4 CANISTERS	0.1 - 0.26
AT THE SITE FENCE	<0.0025

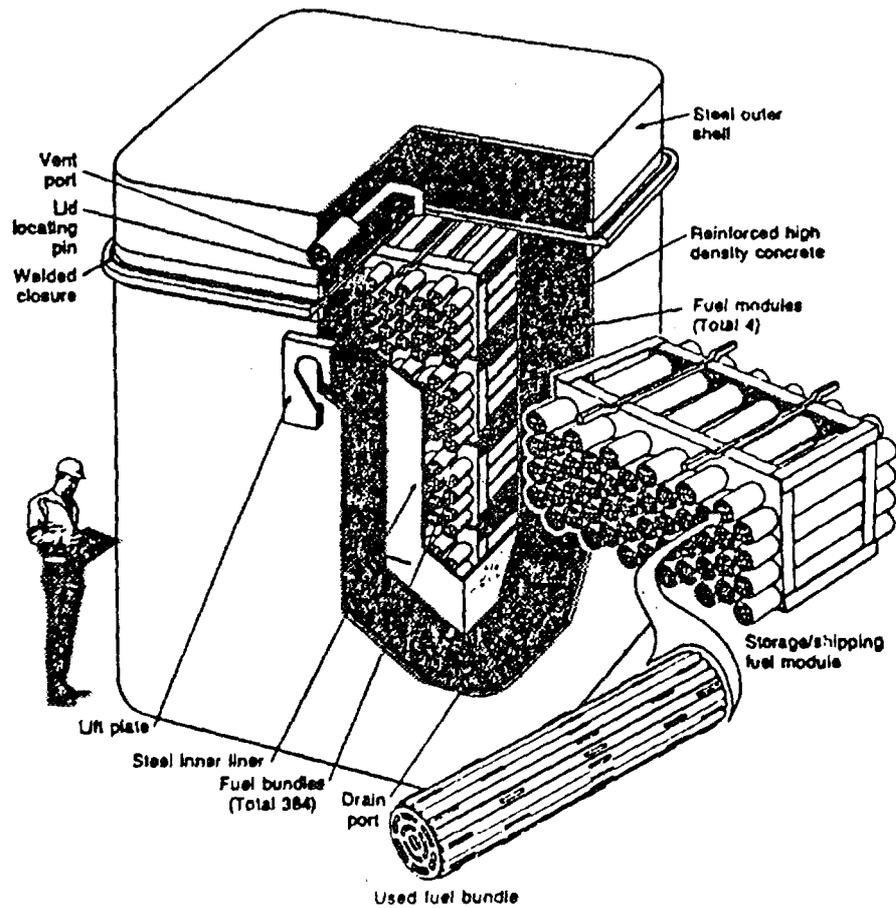


FIG. 6 Ontario Hydro's spent fuel dry storage container (approximate dimensions — height: 3.6 m; front: 2.1 m; side: 2.4 m; gross weight (including fuel): 63 Mg).

3gms. and the moisture in each failed fuel is less than 4 gms. Thus for an expected 0.1% fuel failure rate and a BWR storage cask containing 52 fuel assemblies (7X7), the total moisture in the storage medium will be about 16 gms. This amount of moisture contains about 0.4 moles of oxygen which is expected to convert 60 fuel pellets to U_3O_8 if all oxygen was consumed only by UO_2 [8]. However the limited inventory of moisture is expected to be consumed by the clad and the cask internal materials also and therefore this small amount of moisture is not expected to pose much problem.

As can be seen from the commissioning and initial operation of the Pickering dry storage programme with DSCs (described in section 7.2), on the lines similar to that of LWR storage practice, a lot of emphasis has been placed on moisture removal for storage of CANDU fuel in helium [14].

8.2 Importance of Moisture Control For Storage in Air

In the fuel dry storage in air, the phenomena of oxidation of UO_2 is not completely avoided, but as the oxidation rate will be very slow due to low fuel temperature, it is expected to be within acceptable levels for the expected storage time. The moisture in the cask is however kept to a minimum as the formation of the very low density hydrated uranium oxides formed due to oxidation by moist oxygen could hasten the damage to clad due to high fuel swelling. The moisture content in the dry air and water in the cask is therefore kept to negligible quantities.

8.3 Significance of Orientation of Bundles in Moisture Removal

The Canadian dry storage practice in concrete canisters indicates that the orientation of bundles inside the cask plays an important role especially with regard to moisture removal procedures. This has been further emphasised during the design stage of CANDU-3 where cylindrical basket was planned as the receptacle for storage of used fuel in the storage bay itself so as to avoid the handling operation of tilting and transfer of bundles from trays to baskets[12]. In the case of Dry Storage Container (DSC) where the bundles are stored in horizontal orientation, vacuum drying has been used for removal of moisture. As can be seen from the commissioning and initial operation of the Pickering dry storage programme with DSCs, the 55 litres of water left in the cask after draining was removed by vacuum drying operation in different stages which lasted for almost 5 days in total.

To study the effect of bundle orientation on water hold-up by bundles, an experiment was conducted at Hall-7, BARC. Two wire wrap bundles were immersed in water for about 60 minutes and then taken out. One of the bundles was kept in horizontal orientation and the other in vertical orientation. Water was allowed to drain from the bundles for about 20 minutes. The water trapped inside the fuel bundles (in the gap between the fuel pins) was flushed out into polythene bags by passing compressed air through the bundle. The quantity of water trapped in the bundle kept in vertical position was about 0.2-0.3 cc whereas in the bundle kept in horizontal position, it was about 10 cc. This clearly indicates that the drying process with bundles in horizontal position has to be for a greater moisture removal capacity.

9.0 CONCLUSION

The important conclusions that can be drawn from the review of literature on dry storage of spent fuel and which are applicable to the PHWR dry storage programme are :

1. The number of defective fuel pins in a storage cask will be very few and therefore the amount of oxygen present in the cask would be sufficient to completely oxidise the exposed UO_2 in defective fuel pins. Hence, the rate of oxidation is kept very low by having a limit on maximum fuel temperature to ensure that the total oxidation of exposed UO_2 in defective fuel pins is within acceptable levels for the desired storage time.
2. Moisture in storage cask is kept to minimum possible levels as the potential to cause loss of fuel integrity is more for moist storage medium than that for dry storage medium.
3. Nitrogen can be used as the storage medium since its heat transfer characteristics is comparable to that of air. This would also reduce the concentration of the oxidising agents in the storage medium.

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APPENDIX - 1

COMPARISON OF HEAT REMOVAL CAPACITY OF GASES USED FOR DRY STORAGE OF SPENT FUEL

Convective heat transfer coefficient for horizontal cylindrical surfaces

$$h = \frac{Nu \times k}{d}$$

- h - Convective Heat Transfer Coefficient (W/m²)
- Nu - Nusselt Number
- k - Thermal Conductivity (W/m^{°K})
- d - Diameter of Tube (m)

$$Nu = C (Gr. Pr)^m$$

- Gr - Grashoff Number
- Pr - Prandtl Number
- C,m - Constants

$$Gr = \frac{Q_w g \beta (T_w - T_\infty) d^3}{\nu^2}$$

- g - Acceleration due to Gravity
- β - $1/T_f$
where $T_f = (T_w + T_\infty) / 2$
- T_w - Temperature of Clad , °K
- T_∞ - Temperature of Gas , °K
- Q_w - Heat Flux, W/m²
- k - Thermal Conductivity of Gas
- ν - Kinematic Viscosity of Gas

$$h \propto (Gr. Pr)^m$$

AT 100° C TEMPERATURE (T_f)

- $h_{\text{helium}} / h_{\text{air}} \approx 2.0$**
- $h_{\text{nitrogen}} / h_{\text{air}} \approx 0.96$**
- $h_{\text{argon}} / h_{\text{air}} \approx 0.70$**

