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Behavior of Spent Nuclear Fuel in Water Pool Storage

by
A. B. Johnson, Jr.

September 1977

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Battelle
Pacific Northwest Laboratories

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SUMMARY AND CONCLUSIONS

Storage of irradiated nuclear fuel in water pools (basins) has been standard practice since nuclear reactors first began operation ∿34 years ago. Pool storage is the starting point for all other fuel storage candidate processes and is a candidate for extended interim fuel storage until policy questions regarding reprocessing and ultimate disposal have been resolved.

This report assesses the current performance of nuclear fuel in pool storage, the range of storage conditions, and the prospects for extending residence times. The assessment is based on visits to five U.S. and Canadian fuel storage sites, representing nine storage pools, and on discussions with operators of an additional 21 storage pools. Spent fuel storage experience from British pools at Winfrith and Windscale and from a German pool at Karlsruhe (WAK) also is summarized.

At the end of 1976 there were ∿8700 power reactor fuel bundles in storage in U.S. pools. Approximately 90% of the bundles have Zircaloy cladding; the remainder have stainless steel cladding. Approximately 70,000 Zircaloy-clad bundles (∿50 cm long) were stored in Canadian pools at the end of 1976.

Maximum pool residence for Canadian fuel is 14 years. Zircaloy-clad U.S. fuel has been in pool storage up to 18 years. Experimental stainless-clad fuel has been stored up to 12 years; commercial stainless-clad fuel has been stored up to 7 years; unirradiated stainless steel fixtures have been exposed in pools up to ∿20 years without evidence of degradation Maximum burnups for stored commercial fuel are ∿33,000 MWd/MTU for both Zircaloy- and stainless-clad fuel.

Perceptions regarding the status of the stored spent fuel are based principally on visual observations during fuel handling operations and on visible portions of the bundles during storage. Radiation monitoring of water and air in pool storage areas also is conducted to detect evidence of radiation releases from the stored fuel.

The results of the survey indicate that pool operators have not seen evidence that stainless- or Zircaloy-clad uranium oxide fuel is degrading during pool storage, based on visual examinations and radiation monitoring.

Irradiated Canadian Zircaloy-clad fuel was returned to a reactor after up to 10 years of pool storage, with satisfactory performance. Shippingport fuel was removed from pool storage to a hot cell inspection in air after 4 years in pool storage. There was no visual evidence of degradation and no radiation releases occurred.

Mechanical damage to spent fuel during reactor discharge and fuel handling in the pools is minimal. The number of incidents where fuel was dropped during fuel handling operations appears to have been less than a dozen cases in 1974 to 1976. Only two cases were identified where fuel damage resulted in breached cladding.

Several hundred fuel bundles having rods which developed cladding defects during reactor exposures are in pool storage. Radioactive gases were expelled to the reactor coolant and therefore are not released from the reactor-induced cladding defects during pool storage. However, non-gaseous fission products are released to the pool water. Steady-state radioactivity concentrations in pool water can be maintained in the range 10^{-3} to 10^{-4} μCi/ml with ion exchange and filtration. Higher values (up to $\sim$0.5 μCi/ml) occur during fuel discharges at reactor pools. Spent fuel with defective cladding has been stored, shipped and reprocessed, frequently on the same basis as intact fuel.

The range of storage conditions in fuel pools is outlined below:

- <u>Water Chemistries</u>

  BWR and ISFSI(a) pools:
    Oxygen-saturated deionized water
  PWR pools:
    Oxygen-saturated deionized water + $\sim$2000 ppm boron as boric acid

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(a) Independent Spent Fuel Storage Installation; the only U.S. ISFSI pools which now store spent fuel are GE-Morris and Nuclear Fuel Services.

- Temperature Range

  70 to 120°F (20 to 50°C), bulk water temperatures

  Pools with adequate heat exchanger capacity maintain temperatures below 100°F, even with freshly-discharged fuel; clad temperatures for freshly-discharged fuel are ∿18°F (10°C) above the bulk water temperatures. Mild temperature transients, within the range cited above, have occurred in pools during temporary shutdown of heat exchangers.

- Materials

  Pool walls--painted concrete, stainless steel, fiberglass

  Fuel canisters and racks--stainless steel or aluminum alloys

  Grapples and hoists--stainless- or chromium-plated steel

Detailed, systematic examinations of fuel bundle materials have not been conducted specifically to define storage behavior, because of the expectation that the fuel would be reprocessed after relatively short pool residence. Also, there is minimal reason to expect that the corrosion-resistant fuel bundle materials would degrade in the relatively benign storage environments over the expected storage period. Over the range of pool storage experience cited above, there have been no observations which raise concerns. However, it is not now clear how long pool storage of spent fuel may be extended. If storage times of the spent fuel inventory are expected to extend into the 20-to-100-year time frame, there is an increasing incentive to determine whether any slow degradation mechanisms are operative.

Further assurances regarding fuel cladding integrity can be based on selected destructive exams of spent fuel having a previous exam history, which defined the results of the reactor exposure. Also, periodic visual and non-destructive surveillance of selected stainless- and Zircaloy-clad bundles can provide a systematic, sustained approach to verify the integrity of the spent fuel inventory. Such an approach, of limited scope, has in fact begun in Germany (Karlsruhe). The inspections also should include fuel having reactor-induced defects. Unless evidence of degradation develops in exploratory investigations, a surveillance program involving large numbers of bundles is not justified.

To define certain aspects of long-term (20-to-100-year) spent fuel and pool equipment integrity, some laboratory investigations may be useful. Any detailed fuel investigations and laboratory studies should consider the action of possible degradation mechanisms on either interior or exterior cladding surfaces and on lifting members such as fuel bundle bails. Cladding stresses are not expected to be high, but whether they are sufficient to participate in certain slow degradation mechanisms is not clear. Pitting or other localized corrosion, particularly of stainless steel, cannot be ruled out by present levels of inspection, again in regard to very long exposures.

Based on current experience and on an assessment of the relevant literature, prospects are favorable to extend storage of spent nuclear fuel in water pools, recognizing the following considerations:

- Zircaloy-clad fuel has been stored satisfactorily in pools up to 18 years; stainless-clad fuel has been stored up to 12 years.
- Low temperatures and favorable water chemistries are not likely to promote cladding degradation.
- There are no obvious degradation mechanisms which operate on the cladding under pool storage conditions at rates which are likely to cause failures in the time frame of probable storage.

Recommendations

- There is sufficient evidence of satisfactory integrity of pool-stored fuel to warrant extending fuel storage times and expanding fuel storage capacities.
- Exploratory examination of selected pool-stored fuel is warranted, particularly if the stored fuel inventory is expected to move into the 20-to-100-year time frame, to define whether slow degradation of the fuel bundle materials is operative. To be effective, the examinations must involve bundles having previous destructive examinations which define the effects of the reactor exposure, followed by substantial pool exposures. Periodic visual and non-destructive surveillance of selected bundles can provide further assurance of sustained fuel bundle integrity.

INTRODUCTION

Storage of spent nuclear fuel in water pools is an interim measure, originally intended to culminate in fuel reprocessing after a few months or years of pool residence. Recently, environmental and political considerations have placed the availability of reprocessing in question, prompting increased emphasis on development of interim fuel storage options. Spent fuel handling and disposal options under consideration include extending basin storage, removal of fuel to interim wet or dry storage in metal containers, final disposal of fuel elements without reprocessing, or eventual reprocessing.

The option to extend basin storage would utilize an existing technology. Figure 1 shows spent fuel bundles stored in stainless steel canisters. The upper ends of the bundles are ∿14 feet from the water surface. The unoccupied stainless steel racks to the right of the canisters are ∿27 feet from the water surface. The technology for water storage of spent nuclear fuel has developed over the past 30+ years. Early in nuclear technology development the attractions of water for storage of irradiated fuel were recognized. Water provides cooling of fuel decay heat, mobility for easy fuel transport, shielding against radiation, visibility for fuel inspection, and containment for non-gaseous radioactivity released from the fuel. Figure 2 shows a spent fuel bundle during transfer from a shipping cask to a stainless steel storage canister. The top of the cask is ∿33 feet from the pool surface; the top of the storage canister is ∿34 feet below the pool surface during the transfer operation. The BWR canister (A) holds nine spent fuel bundles. The PWR canister (B) occupies the same rack space as the BWR canister, but holds only four PWR bundles, which are larger than BWR bundles. Figure 3 shows a BWR storage canister containing nine spent BWR fuel bundles being transported to a storage location.

The current regulatory guidance for fuel storage in U.S. Independent Spent Fuel Storage Installations is based on 10 CFR 30, 40, and 70 and on Regulatory Guide 3.24 and endorses ANSI Standard N-305. Regulatory guidance for pools at nuclear reactors is based on Regulatory Guide 1.13, endorsing ANSI Standard N-210, Part 50.

Early nuclear fuel was aluminum-clad and is therefore not pertinent to predictions of power reactor fuel behavior. Fuel with stainless steel and Zircaloy cladding was irradiated in test reactors beginning in the late 1950's. The first U.S. power reactors were Shippingport (PWR, 1957) and Dresden I (BWR, 1960). The first Canadian power reactor was NPD (Nuclear Power Demonstration, PHWR, 1962). Much of the fuel from the early test and power reactors was reprocessed at the Nuclear Fuel Services plant or at government reprocessing sites. However, some fuel has remained in pools over extended exposures.

The prospects that pool storage may be substantially extended and expanded has prompted numerous questions from licensing bodies, intervenors, etc., regarding the long-term integrity of stored fuel. Concerns center on the prospects that fuel cladding might degrade, which could lead to radiation releases or could compromise future operations such as packaging for dry storage or shipping and handling in reprocessing operations. The nuclear industry must respond to reasonable concerns from a sound technical position. A major aspect of that position is to assess and document the performance of the existing stored fuel inventory.

This report summarizes results of a survey of basin storage experience based on discussions with basin operators and from the literature. The survey comprises a summary of fuel performance in basin storage and identification of the range of basin storage conditions. The storage conditions are analyzed in terms of known material degradation mechanisms.

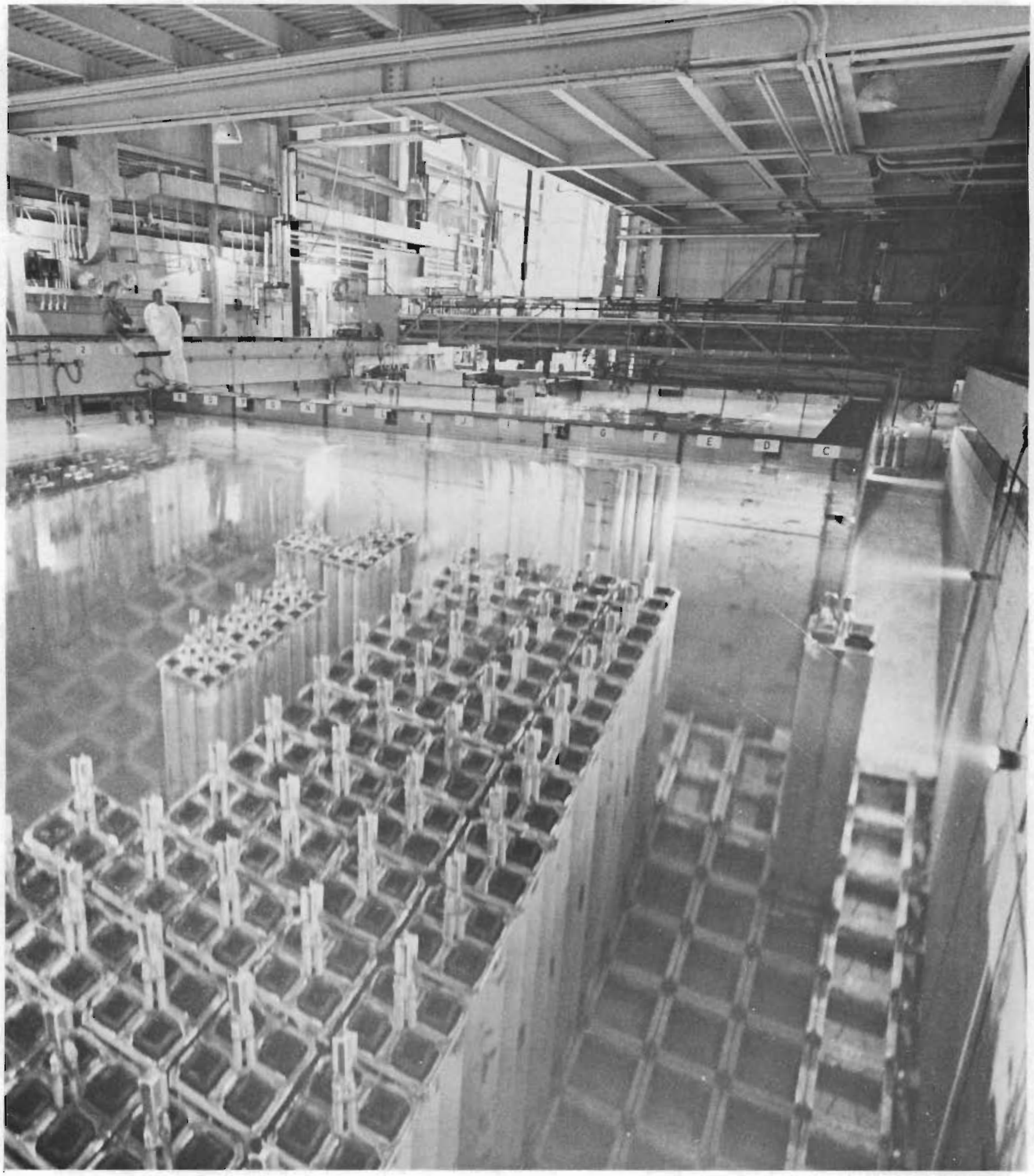

FIGURE 1. G.E. Morris Operation - Fuel Pool - Water Depth, 28.5 ft (8.7m)

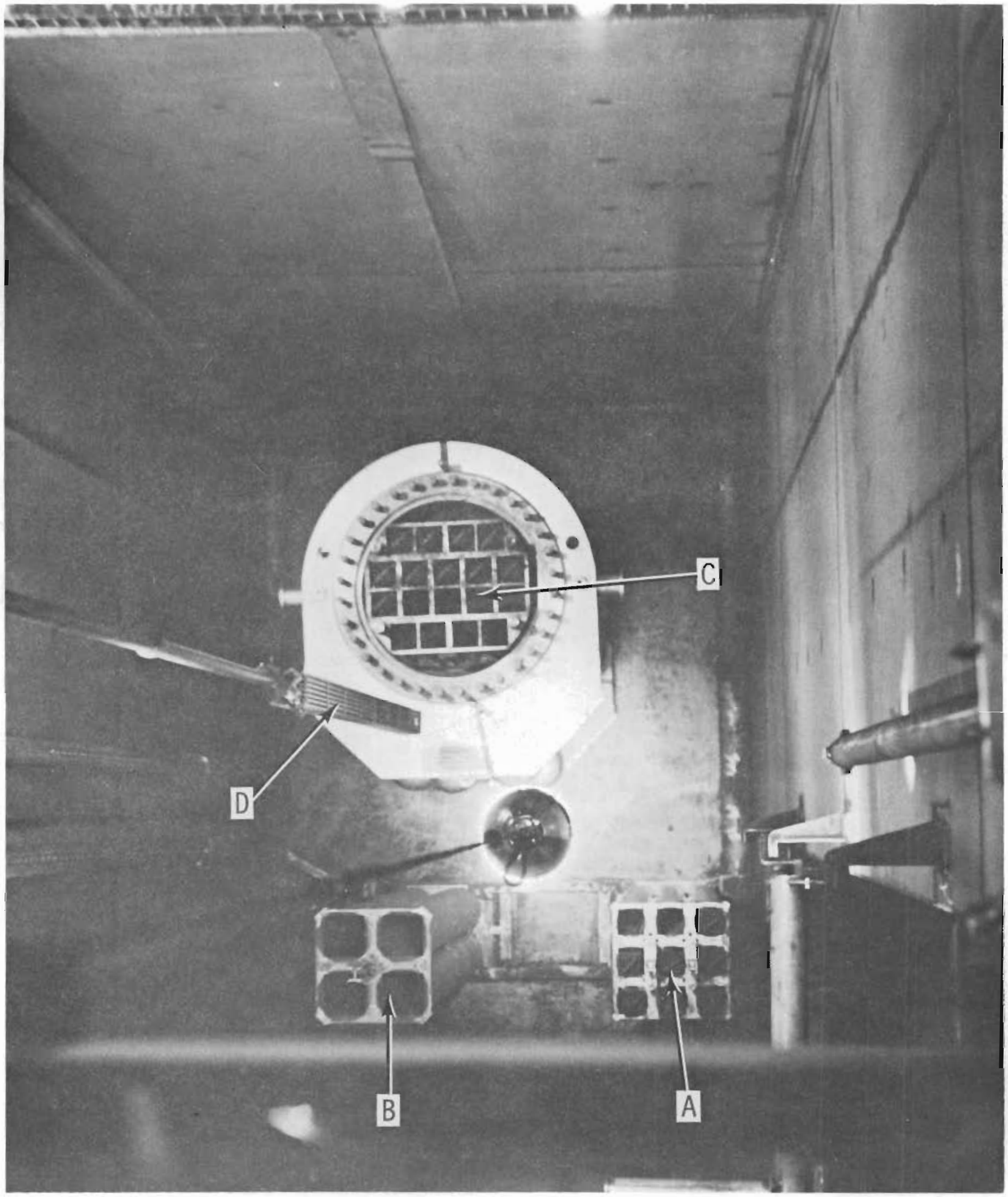

FIGURE 2. G.E. Morris Operation - Spent Fuel Bundle (D) Being Transferred from IF-300 Shipping Cask (C) to BWR Storage Canister (A). A PWR Canister also is shown (B). Water depth, 48 ft. (14.6 m).

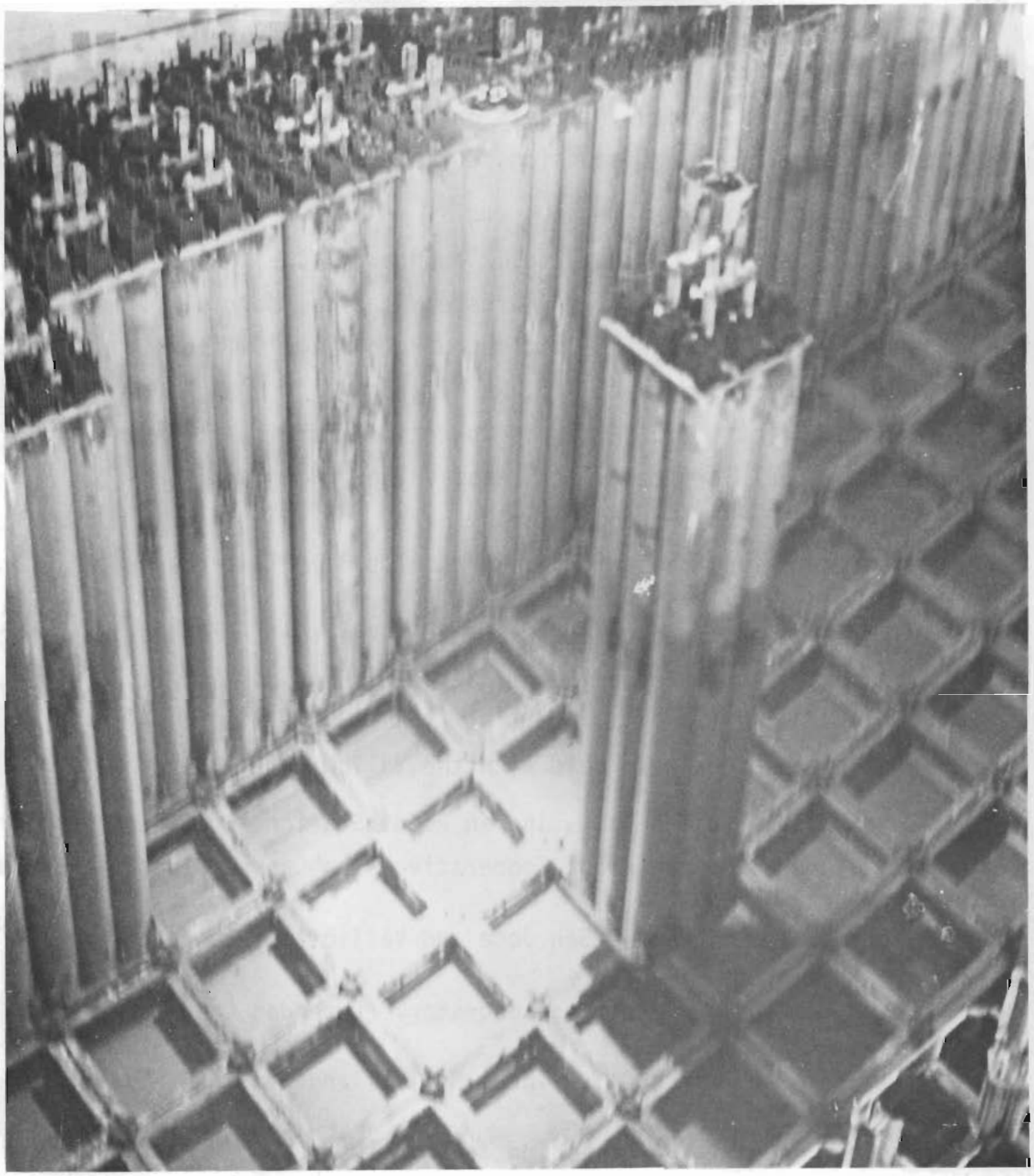

FIGURE 3. G.E. Morris Operation - Fully-Loaded Storage Canister Being Transported to a Pool Storage Location. Water depth, 28.5 ft (8.7 m).

SCOPE OF FUEL POOL SURVEY

The survey involved contacts to a representative cross section of the nuclear industry with interests or direct involvement in pool storage of spent nuclear fuel. Discussions during visits were held with the following:

Atomic Energy of Canada, Ltd., Toronto and Chalk River, Ontario

Commonwealth Edison Co. - Dresden Station, Morris, Illinois

Electric Power Research Institute, Palo Alto, California

Exxon Nuclear Co., Richland, Washington

General Electric Co. - Morris Operation, Morris, Illinois

Nuclear Fuel Services, Inc., West Valley, New York

Ontario Hydro, Toronto, Ontario

Pacific Gas and Electric Co. - Humboldt Bay Plant (Eureka) San Francisco, California

United Nuclear Industries - Hanford N-Reactor, Richland, Washington

Washington Public Power Supply System - Hanford Nos. 1, 2, and 4, Richland, Washington

Telephone discussions were held with the following:

Consolidated Edison Co. - Indian Point Station, Buchanan, New York

Dairyland Power and Light Cooperative - La Crosse Plant, La Crosse, Wisconsin

General Electric Co. - San Jose and Vallecitos Laboratories, California

Idaho National Engineering Laboratory (Expended Core Facility) Idaho Falls, Idaho

Metropolitan Edison Co. - Three Mile Island Station, Middleton, Pennsylvania

Nuclear Regulatory Commission, Washington, D.C.

Southern California Edison Co. - San Onofre Station, San Clemente, California

Westinghouse, Bettis Atomic Power Laboratory (Shippingport), Pittsburgh, Pennsylvania

Yankee Atomic Energy Co. - Connecticut Yankee Plant, Haddam Neck, Connecticut and Yankee Rowe Plant, Rowe, Massachusetts

The survey involved questions on the following topics:

- Date fuel storage facility was placed in service
- Pool size and fuel capacity
- Materials in contact with fuel bundles and basin water
- Pool water chemistry and chemistry control procedures
- Pool and fuel rod temperatures
- Radioactive species and concentrations in pool water
- Fuel rod radiation levels

Characteristics of stored fuel

- Types of cladding
- Maximum burnups
- Longest residence times

Fuel Pool Storage Experience

- Storage experience with spent fuel
- Storage experience for fuel which failed in-reactor
- Mechanical damage during fuel handling operations

Excellent cooperation and detailed responses from the organizations listed above are gratefully acknowledged.

The following section summarizes the current scope of pool storage, the range of storage conditions, and the behavior of the fuel, based on the survey.

NUCLEAR FUEL STORAGE IN WATER POOLS

This section summarizes fuel storage inventories in Canadian and U.S. facilities, including maximum residence times and fuel burnups.

STORED FUEL INVENTORIES

The inventories of nuclear fuel stored in U.S. and Canadian fuel pools in 1976 are summarized in Table 1. The Canadian fuel is Zircaloy-clad. Fuel in U.S. pools includes Zircaloy as the principal cladding alloy, but also includes fuel with stainless steel cladding from both boiling water reactor (BWR) and pressurized water reactor (PWR) electric generating plants. Approximately 90% of the fuel bundles now stored in

TABLE 1. Summary of Canadian and U.S. Fuel Pool Inventories

Country	Number of Bundles
Canada(1)	~70,000[a]
U.S.(2)	8,674[b,c]

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[a] Nominal bundle size(1):
Rods: 50 cm long; 1.5 to 2.5 cm diam.
Rods/bundle: 7 to 37

[b] Includes 2949 bundles discharged in 1976:
1536 BWR; 1413 PWR

[c] Nominal bundle size(2)
BWR: 4.0 m long; 1.25 cm diam.
Rods/bundle: 49 to 64 (Zircaloy); 100 (stainless steel)
PWR: 3.8 m long; 0.95 cm diam.
Rods/bundle: 225 to 289 (Zircaloy); 196 to 225 (stainless steel)

U.S. pools have Zircaloy-2 or Zircaloy-4 cladding. The nominal compositions for the two alloys are: Zircaloy-2, 1.5 Sn, 0.20 Fe, 0.15 Cr, 0.05 Ni; Zircaloy-4, 1.5 Sn, 0.24 Fe, 0.13 Cr, 0.005 Ni. Zircaloy-2 is the principal BWR cladding and Zircaloy-4 is the principal PWR cladding. The behavior of the two alloys is expected to be similar under pool storage conditions. Only three U.S. power reactors currently operate with stainless steel cladding: Connecticut Yankee (PWR, 600 MWe), San Onofre 1 (PWR; 450 MWe), and La Crosse (BWR, 56 MWe). Stainless steel fuel cladding materials are alloys 304 and 348H.

The anticipated fuel storage requirements and storage facilities are summarized elsewhere for Canada(4) and the U.S.(2,5)

MAXIMUM FUEL BURNUPS

Table 2 summarizes maximum burnups on fuel now in storage, reported by those who participated in the survey.

TABLE 2. Maximum Fuel Burnups on Stored Commercial Fuel

Cladding	Reactor	Burnup* MWd/MTU Maximum	Reactor Discharge Date
Zircaloy-2	BWR	25,000	1974
Zircaloy-4	PWR	33,160	1976
Stainless Steel	PWR	33,200	1973
Stainless Steel	BWR	22,000	1975

* Applies to burnup on peak bundle.

Fuel burnup expressed as MWd/MTU is a measure of the power generated by the fuel. On the other hand, radiation damage phenomena generally are correlated in terms of neutron fluence (neutrons/cm^2). A rigorous conversion from burnup (e.g., MWd/MTU) to fluence must account for the physics of the specific fuel and reactor in question. However, the following expression approximates the conversion for a pressurized water reactor (Based on data in WCAP-8837 by H. H. Crain, T. E. K. Caye, and P. J. Sipush, Tables 2-4 and 2-5):

$$1 \text{ MWd/MTU} \approx 1.7 \times 10^{17} \text{ n/cm}^2, \quad > 1 \text{ MeV}.$$

Thus, the estimated neutron fluence corresponding to 33,000 MWd/MTU is 5.6×10^{21} n/cm^2, >1 MeV. Values for BWRs would generally be lower due to lower power densities. Changes in cladding properties, such as increased strength, decreased ductility and metal growth have been reported for Zircaloy(6) and stainless steel.(7) The residual radioactivity in the cladding(8) and the fission product inventories(9) and heat generation rate are relatable to fuel burnup, but they decrease with time after discharge, due to radioactive decay.

FUEL POOL RESIDENCE TIMES

The longest basin exposures of power reactor fuel, identified in the survey, are summarized in Table 3. Canadian Zircaloy-clad fuel from the NPD reactor has been stored in aluminum alloy canisters for 14 years (in 1977) in the NRU reactor basin at the Chalk River Nuclear Laboratories.

TABLE 3. Maximum Fuel Bundle Residence Times in Pool Storage

No. of Bundles	Cladding	Reactor	Date in Pool Storage	Location	Burnups,* MWd/MTU Maximum
25	Zircaloy-2	PHW	1963	NRU/AECL	10,000
1(a)	Zircaloy-2	PWR	1959	ECF/Idaho	6,000~~(b)~~
47	Stainless	PWR	1970	GE/Morris	19,900
∿60	Stainless	BWR	1963(c)	SRP/So.Car.	10,000

(a) Two additional Shippingport bundles are in storage at ECF, one discharged in 1961 and one discharged in 1964.

~~(b)~~ Peak core; average core is 10,860 MWd/MTU, (WAPD 320, October 1969).

(c) Reprocessed in 1975.

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* Applies to burnup on peak bundles.

In 1977, three Canadian fuel bundles were re-irradiated after pool storage for 10, 9, and 5 years, respectively. They are enriched, 28-element bundles. The first irradiations were to relatively low burnups, but at reasonably high power levels. The bundles were re-irradiated for approximately one month with no indications of defects. During the second irradiation, the bundles operated close to Pickering power ratings, (4.2 kw/m, nominal heat rating).

One Shippingport Zircaloy-clad fuel bundle has been stored for 18 years in a stainless steel rack at the Shippingport reactor, and in the Expended Core Facility (ECF) at the Idaho National Engineering Laboratory, where it was transported for metallographic examination of selected rods. In December 1976, after 4 years of pool exposure, a Shippingport bundle was removed to a hot cell and visually inspected in air. No evidence of degradation or radiation release was detected.

Zircaloy-clad and stainless steel-clad UO_2 fuel bundles were irradiated to peak burnups of $\sim$10,000 MWd/MTU in the General Electric Company Vallecitos Boiling Water Reactor (VBWR). The bundles are either 3 by 3 or 4 by 4, 0.563 in. O.D., $\sim$3 feet long. Sixty-three Zircaloy-clad assemblies discharged from VBWR in 1963 were stored in the pool at that site until 1964/65, and were then shipped to the Savannah River Plant, (SRP) and are still stored there in a water pool.

Approximately 60 stainless-clad assemblies also were discharged and stored at VBWR until shipment to SRP in 1964/65. Reprocessing of the stainless-clad fuel was completed in 1975.*

There have been no problems with this Zircaloy- or stainless-clad fuel in storage, shipping and handling.

The behavior of fuel stored in Canadian and U.S. water pools is summarized in the following section.

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*Approximately 47 additional stainless-clad VBWR bundles which developed defects during the reactor exposure are stored in closed cans. The fuel bundles are covered by water inside the cans, but are isolated from pool water. The canned fuel is scheduled for reprocessing late in 1977.

FUEL PERFORMANCE DURING POOL STORAGE

SURVEILLANCE METHODS

Assessment of spent fuel behavior in pool storage is based principally on visual observations and on radiation monitoring of the pool water. Many fuel bundles are relocated within the pool or are removed from storage for shipment after substantial pool residence, providing opportunities for visual inspection (Figure 2). Fuel rods sometimes are subjected to detailed nondestructive and destructive examinations, to define the results of the reactor exposure, but the exams generally occur after short pool residence. Successful re-irradiation of Canadian fuel after up to 10 years of pool residence and inspection of Shippingport fuel in a hot cell after 4 years in pool storage provide significant confirmation of sustained integrity of Zircaloy-clad fuel (see preceeding section).

Radiation levels are monitored by periodically sampling pool water and by continuously monitoring air above the pool. In reactor pools, the radioactivity concentrations increase significantly while fuel is being discharged from the reactor, (for example, from $\sim 10^{-5}$ µCi/ml to $\sim 10^{-2}$ µCi/ml at the Humboldt Bay reactor) due to mixing of reactor and basin waters. In ISFSI pools (currently, GE-Morris and NFS), radiation levels have correlated with incoming fuel shipments, but are generally more stable than the levels in reactor pools. If significant cladding failures were to occur during pool storage, they almost certainly would be signaled by deviations from the established radiation history for a given pool.

In the survey, basin operators were asked to summarize experience regarding the following aspects of spent fuel behavior in pool storage:

- Evidence of fuel bundle material degradation during pool storage.
- Mechanical damage prior to and during pool storage.
- Behavior of fuel which failed in the reactor.

Fuel pool operator responses are summarized below.

POOL OPERATOR OBSERVATIONS ON FUEL BUNDLE CORROSION

Responses from Atomic Energy of Canada and Ontario Hydro indicated that there has been no evidence of significant corrosion or other chemical degradation on the Zircaloy-clad bundles now in storage in Canadian pools (Table 1). This includes bundles which have up to 14 years of pool residence (Table 3). Some of the early bundles were inspected by removal from the canisters occasionally during the 14-year residence without noting degradation. The Canadian experience offers substantial assurance that Zircaloy-water interactions are not problematic, based on visual observations and radiation monitoring.

Basin operators representing 20 U.S. pools (see page 10) also indicated that they have not seen visual evidence or radiation releases which portend fuel cladding degradation during water pool residence.

The only case of substantial fuel element degradation during pool exposures involved N-Reactor fuel stored in the NFS pool prior to reprocessing. The N-Reactor fuel is Zircaloy-clad metallic uranium. The Zircaloy cladding on a fraction of the elements (1 to 2%) was damaged when the fuel was discharged from horizontal process tubes into an underwater receiving chute.* Water reactions with the areas of exposed metallic uranium during shipping and in storage at NFS appear to account for the fuel degradation. All N-Reactor fuel which was in storage at NFS has been reprocessed. In contrast to the behavior of the damaged metallic fuel, the uranium oxide pellets from a few damaged commercial power reactor rods stored at NFS appeared to be inert to the pool water.

A few cases of material degradation have occurred in pools at experimental facilities. One case involved unusual water chemistry ($\sim$400 ppm NO_3^- and 360 to 750 ppm Cl^-), which caused extensive corrosion of aluminum fuel element storage hangers.(10) At another site, severely-sensitized stainless steel rods failed by intergranular corrosion in a

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*By contrast, fuel bundles at commercial power reactors are carefully handled individually, similar to the method shown in Figure 2.

moist cover gas, and in pool water(11). At a third site, two Zircaloy-clad metallic uranium fuel elements partially disintegrated during pool storage, in high-purity water (<0.5 μmho/cm) at the Savannah River Plant, South Carolina. Cladding on both elements became defective during the reactor exposure. The failure mechanism during pool storage appears to be identical to that which occurred on the N-Reactor fuel, involving water reaction with the uranium metal. These examples, while not characteristic of commercial fuel, illustrate the importance of maintaining satisfactory pool chemistry and fuel bundle metallurgical conditions which are compatible with the storage environment. They also emphasize that in contrast to metallic fuel, the oxide fuel appears to have far superior durability in pool storage, including numerous rods which developed defects during the reactor exposure. Finally, they illustrate that even if fuel failures occur, pool storage technology is available to deal with them. The subject of storage behavior of defective fuel is treated in the following section.

HANDLING FAILED FUEL IN BASIN STORAGE

Fuel with cladding defects which developed in the reactor have had extended residence in water pools without major impacts on pool operation. Recent concerns have been voiced regarding the status of defective fuel in storage. This section summarizes perceptions from pool operators regarding storage characteristics of defected fuel. It also provides a perspective regarding the types of reactor-induced failures which are stored.

Procedures for Handling Defective Fuel

Some special procedures have been developed to deal with failed fuel, including both fabrication of underwater hoods intended to collect radioactive gases if fuel were to leak in the pool, and special canisters to isolate failed fuel from pool water. In some instances these procedures prove useful. However, the vast majority of failed fuel does not require special handling and is stored in the same manner as intact fuel. Two aspects of the defective fuel account for its favorable storage characteristics. First, when a fuel rod perforates in-reactor, the radioactive gas inventory is released to the reactor primary coolant.

Therefore, upon discharge, little additional gas release occurs. Only if the failure occurs by mechanical damage in the basin are radioactive gases released in detectable amounts, and this type of damage is extremely rare (see Table 5). The second favorable aspect is the inert character of the uranium oxide pellets in contact with water. This has been demonstrated in laboratory studies(12) and also by casual observations of pellet behavior when broken rods are stored in pools.

Some pools are equipped with vacuum cleaners to remove radioactive debris from the bottom of the pool.

The case histories below provide additional insights to handling of defective fuel in pool storage.

Nuclear Fuel Services Experience with Defective Fuel

The principal source of failed fuel in the NFS fuel pool was the Zircaloy-clad metallic uranium N-reactor fuel mentioned earlier. During the period 1968 to 1970 over 150 failed N-reactor fuel elements were stored in the pool. Concentrations of radioactive species in the pool water rose from normal levels near 10^{-3} to just below 10^{-2} μCi/ml.

In November 1971 the last of the existing fuel inventory in the NFS pool was reprocessed. The pool was drained and the pool bottom (painted concrete) was vacuumed and scrubbed with brushes. The radiation levels on the bottom of the pool were reduced to ∿150 mR/hr, including some activity from radiation incorporated in corrosion products on the aluminum storage racks. After the pool was decontaminated and refilled, the pool water radioactivity levels returned to between 10^{-3} to 10^{-4} μCi/ml. Pool water purification consisted of ion exchange and filtration.

The current fuel inventory stored in the NFS pool is 165 tonnes (756 assemblies), including over 100 bundles with rods which leaked in-reactor. The lower limit of radioactivity in the pool water with filtration and ion exchange is 8×10^{-4} μCi/ml. When circulation of water to the cleanup facilities was shut off for one month, the concentrations rose to 2 to 3×10^{-3} μCi/ml. The principal radioactive species are ^{134}Cs, ^{137}Cs, ^{60}Co, and ^{65}Zn. Tritium is a minor species. Cesium isotopes account for 95 to 99% of the gamma activity.

Humboldt Bay Reactor, Failed Stainless Steel Fuel

In early BWR operation, numerous fuel failures occurred in stainless steel fuel cladding, due to stress corrosion cracking in the oxygenated BWR primary coolants.(13) This problem developed on the first Humboldt Bay core (1963). The stainless steel cladding began to fail after ∿2000 MWd/MTU, involving at least one fuel pin failure per bundle. The fuel was stored uncanned in the Humboldt Bay pool. In some cases, ruptured fuel rods were observed. Radioactivity concentrations in the basin water were normally 10^{-5} μCi/ml, but rose to $\sim 10^{-2}$ μCi/ml during the discharge of the failed fuel. The primary activity was due to ^{65}Zn, from activation of corrosion products from Admiralty Brass heat exchangers. The activity was incorporated in the crud layer on the fuel rod surfaces. Current radioactivity in the pool water is $\sim 10^{-3}$ μCi/ml. The soluble species are principally ^{134}Cs and ^{137}Cs, but a major fraction of the activity comes from particulates from reactor crud species, principally ^{60}Co and ^{54}Mn. The ^{65}Zn contribution has diminished since the Admiralty Brass heat exchangers were replaced.

Over a period of five years, 270 bundles with stainless steel cladding were shipped to the NFS plant for reprocessing. Approximately 190 of the bundles with failed rods were placed in closed containers just prior to shipping. While increasing manpower and costs, handling of the canned fuel in the basin, in shipping, and reprocessing was accomplished without substantial problems.

Storage of Defective Zircaloy-Clad Power Reactor Fuel

Operators at several reactors have discharged, stored, and/or shipped relatively large numbers of Zircaloy-clad fuel which developed defects during reactor exposures, e.g., Ginna, Oyster Creek, Nine Mile Point, and Dresden units I and II. Several hundred Zircaloy-clad assemblies which developed one or more defects in-reactor are stored in the GE-Morris pool without need for isolation in special cans. Detailed analysis of the radioactivity in the pool water indicates that the defects are not continuing to release significant quantities of radioactivity.(20) Normal radioactivity concentrations in the Morris pool water are

$\sim 3 \times 10^{-4}$ μCi/ml which is near the maximum desired concentration for occupational exposure considerations, in bathing, culinary uses, ect. The radioactivity concentrations rose to 2×10^{-3} μCi/ml during a month when the water cleanup system was removed from service.

Canadian handling procedures for Zircaloy fuel with defects include storage in separate cans or in sealed cans.

Fuel Defect Mechanisms

The causes of fuel defects during reactor exposures have been summarized by Locke,(14) Robertson,(15) and Bobe.(16) They include manufacturing deficiencies such as faulty welds, wall cracks and impurities in the fuel cladding; external factors such as handling damage, fretting and a few cases of cladding corrosion, principally crud-induced failure of Zircaloy cladding and stress corrosion cracking of stainless cladding in BWRs. Internal causes include fission product attack, pellet-clad interactions and fuel densification.

Fuel Failure Statistics

Some fuel failure types have resulted in occasional epidemics, involving numerous fuel failures in several reactors. Examples include internal hydriding and fuel densification. In several reactors, large fractions of the fuel bundles were removed to pool storage after low burnups ($<$5000 MWd/MTU) due to fuel failures. Most of the failure mechanisms have been eliminated. Locke(14) cites failure rates of one rod in 100 rods per reactor year in some reactors during some of the epidemics, but indicates that current technology is capable of operating at one failure in 10^4 rods per year. The number of fuel rods in 1000-MWe BWRs and PWRs is 40,000 - 50,000. Based on the failure rate cited above, the expected defect rate is 4 - 5 rods per year per 1000-MWe plant. While several compilations provide data on fuel failure statistics,(14-16) the total inventory of failed rods in U.S. pools can only be estimated. The number of bundles with one or more failed rods probably is 1400 to 1600, based on data compiled by C. W. Funk, Westinghouse Hanford Engineering Development Laboratory. Of these, the estimated number which have been reprocessed is 200 to 400. The failure rate for Canadian fuel is 0.03%,(1) suggesting that $\sim$500 to 600 rods with defects are now in storage in Canada. The defects have not caused problems in fuel handling or storage.

Types of Defects

The principal types of defects in stored fuel are:

- pinhole defects in welds, hydride blisters, etc.
- longitudinal and circumferential cracks due to hydriding and pellet-clad interactions
- large breaks in the cladding, occasionally with cladding sections missing

Pinholes and small cracks appear to be the most common defect types; broken rods occur infrequently.

Significance of Defective Fuel Behavior in Pool Storage

The experience cited above indicates that nuclear fuel with reactor-induced defects can be handled, shipped, stored, and reprocessed satisfactorily. This implies that substantial degradation of pool-stored fuel could also be tolerated without compromising pool safety. However, it appears prudent to maintain surveillance of fuel bundle materials as pool residence times increase, to determine whether substantial deterioration occurs on defective or intact spent fuel. Degradation rates almost certainly would be low under pool storage conditions, providing time to develop procedures to deal with problems (e.g., removal to dry storage). Current experience suggests that deterioration of stored fuel is minor. Canadian investigators have projected probable pool storage lives of 100 years for Zircaloy-clad fuel.(4)

MECHANICAL DEGRADATION OF FUEL BUNDLE MATERIALS DURING FUEL HANDLING OPERATIONS

The principal spent fuel handling operations are outlined below:

- Discharge from reactor
- Transfer of fuel bundles to fuel storage canisters
- Transfer of canisters to storage racks and infrequent relocation of racks within the pool
- Transfer of fuel bundles from fuel pool canisters to shipping casks
- Cask shipment by truck or rail
- Transfer from cask to canister at an ISFSI pool (Figure 2), followed by canister placement on storage racks (Figure 3) and infrequent relocation of canisters within the pool.

Eventually the fuel will be removed from reactor pools or ISFSI pools to one of the following options(3-5);

- Reprocessing
- Packaging for interim wet or dry storage
- Processing and packaging for final storage, either as an intact bundle or in an altered form inside a permanent storage container.

Definition of the probable chemical and mechanical states of the cladding when the processing options outlined above occur should be a significant aspect of any pool storage surveillance program.

The mechanical state of cladding upon discharge from the reactor is well-known as a function of reactor exposure for Zircaloy(6) and stainless steel.(7) Neutron irradiation increases the strength and lowers the ductility of all fuel bundle materials. Hydrogen absorption in metals also tends to lower their ductilities. Hydrogen concentrations in irradiated Zircaloy cladding are typically 100 to 150 ppm in BWR cladding at the end of the reactor exposures.(17) In spent PWR cladding, hydrogen concentrations are ∿50 wt ppm, average, and 115 wt ppm, maximum, at burnups of 35,000 to 40,000 MWd/MTU.(18) Uniform hydrogen concentrations in the ranges cited for BWR and PWR cladding are well within the tolerable range for reactor operation and also have not shown evidence of operational problems during fuel handling at fuel pool temperatures. Due to low solubilities and relatively high mobilities of hydrogen in stainless steels, the hydrogen contents of stainless-clad spent fuel under basin storage conditions is not known, but the concentrations are expected to be less than one ppm.

While the basis is available in the literature to define the mechanical state of the cladding as a function of hydrogen content and neutron fluence, operational problems from these sources have generally been minor in-reactor and in pool storage. The combined effects of radiation damage and hydrogen absorption from the external (coolant) surface have not imposed operational problems to either stainless steel cladding under PWR conditions or to Zircaloy cladding under BWR and PWR conditions. Hydrogen from moisture impurity in the uranium oxide fuel has caused numerous in-reactor failures of Zircaloy-clad fuel. Stress corrosion cracking caused numerous failures on stainless steel cladding in BWRs. However, the La Crosse BWR has continued to use stainless steel cores since startup in 1969.

More pertinent to this report is the actual behavior of fuel bundles in the pool storage operations to which they are subjected. Based on the survey and information in reactor incident reports, fuel bundle damage in fuel handling operations was assessed.

Pool Operator Observations on Mechanical Damage

Pool operators contacted in the survey were asked to characterize the type and incidence of mechanical damage which occurs during fuel handling operations.

The description by one pool operator that fuel bundles are handled "as though they are fine crystal" appears to apply in general to fuel handling in the pools. Except for a few fuel handling accidents (next section), the mechanical state of the cladding was fully satisfactory to endure all movements within the pool.

Power reactor fuel stored in pools functioned satisfactorily in the handling and shearing operations in the NFS fuel reprocessing plant, based on discussions with J. P. Duckworth of the NFS staff. This included fuel with the following burnups and pool residence times (Table 4):

TABLE 4. Burnups and Pool Residence Times for Reprocessed Fuel[a]

Zircaloy:	Maximum burnup	- 23,000 MWd/MTU	(BWR)
	Maximum pool storage	- 5 y	(PWR)
Stainless Steel:	Maximum burnup	- 32,000 MWd/MTU	(PWR)
	Maximum pool storage	- 5 y	(PWR)
Inconel 600:	Maximum burnup	- 17,000 MWd/MTU	(BWR)
	Maximum pool storage	- 2.5 y	(BWR)

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[a] Reprocessed at Nuclear Fuel Services Plant, West Valley, New York, 1967-1971.

Fuel bundles have been partially disassembled and/or reconstituted in some pools. This generally appears to occur without problems, and reconstituted bundles are returned to the reactor core. However, at NFS, some fuel rod breakage occurred during attempts to remove rods from fuel bundles by pulling. Pushing was a more satisfactory procedure to avoid fuel rod breakage.

Summary of Fuel Handling Accidents From Reactor Incident Reports

When fuel is damaged during handling operations at reactor sites, the circumstances are reported to the Nuclear Regulatory Commission. Review of Abnormal Occurrence Reports for 1974[a] through 1976 identified nine fuel damage incidents at power reactors (Table 5). Assuming that the fuel handling incidents reported in Table 5 represent most if not all of those which occurred, the incidence of damage appears to reflect minor degradation of fuel bundle materials due to fuel handling damage at pools. In the majority of cases (seven of nine) no breached cladding

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[a] One report applies to late 1973.

TABLE 5. Summary of Incidents Involving Mechanical Damage to Irradiated Fuel Bundles - 1974-76(a)

	Date	Reactor	Operation	Event	Consequence	Probable Cause	Corrective Action	Reference Docket No.
1.	March 31, 1976	Brunswick (BWR)	Fuel bundle transfer from spent fuel storage location to fuel prep machine.	Fuel bundle fell from fuel prep machine to a horizontal position across top of fuel storage racks.	Building evacuated. No radiation release detected. No visible damage to bundle. Bail on another bundle was bent by falling bundle.	Fuel bundle misaligned in the upper guide; bundle not properly seated in prep machine.	Channeling/dechanneling. Revised to include two independent visual verifications of fuel bundle nose piece seating in fuel prep machine.	50324 (letter report dated April 16, 1976)
2.	November 8, 1975	Crystal River (PWR)	Fuel bundle transfer from shipping container to inspection station.	Fuel bundle fell ~ 5 ft to pool floor.	Area evacuated. No release of radioactivity. Some fuel rods were bowed; none appeared to be ruptured. Some spacer grids were broken. Lower end fitting was bent. Some welds on fuel handling tool cracked.	Wire cable for fuel handling cable pulled out of swaged fitting.	(Preliminary) Eliminate swaged connections or develop fuel handling tool which avoids use of slings.	50302-211 or -209
3.	June 10, 1975	Duane Arnold (BWR)	Fuel bundle movement from reactor core to spent fuel pool.	Fuel bundle dropped ~ 30 ft from refueling into core, impacting on the top of another fuel assembly, and coming to rest at 70° angle against reactor vessel wall.	Refuel floor evacuated and door locked. No airborne activity was observed after drop or while dropped bundle was moved. Chemical analysis indicated no increase in iodine or other fission products in reactor water. Underwater TV inspection indicated deformation of lower tie plate on dropped bundle. Impacted bundle had some bail and channel damage and showed momentary release of activity when moved, indicating some fuel rod damage.	Inadequate method to verify proper grapple-fuel engagement.	Bundle placed in storage can. TV monitor used to verify fuel grapple seating until design change completed.	50331-518
4.	June 5, 1975	Humboldt Bay (BWR)	Fuel bundle movement from transfer basket to pool storage location.	Channeled bundle disengaged from grapple - fell 6 ft to pool bottom, tipped over; fell into 10-ft deep cask pit.	Personnel were evacuated; no radioactive gases were detected; the fuel channel was bent; tie rods and/or tie rod keeper sheared, loosening some rods from bundle.	Bundle probably not grappled properly or checked before movement.	Instituted visual monitoring to assure proper closure of grapple hooks.	50123-326
5.	June 3, 1975	La Crosse (BWR)	Relocation of fuel bundles in core.	Spacer grid and external band dislocated and damaged by upper edge of fuel assembly shroud.	Underwater TV inspection suggested fuel rods not damaged. No evidence of radiation release. Damaged parts not repairable.	Tensiometer locked by operator, preventing indication that fuel bundle was binding.	Locking bar eliminated.	50409-230 and -236
6.	June 2, 1975	Surry (PWR)	Refueling.	Only two locking fingers on fuel handling crane engaged, allowing bundle to pivot against another fuel assembly.	Damage to two hold-down springs.	--	--	50281 Special Report SR-52-75-02
7.	May 6, 1975	Turkey Point (PWR)	Fuel bundle removed from spent fuel pit to place into reactor core.	Spent fuel pit side lifting frame struck fuel assembly, causing damage to spacer grid and to two fuel rods.	No radioactivity was released. Fuel rods not breached.	Procedural deficiency.	Procedures modified to correct lifting frame contact with fuel bundle.	50251-352
8.	September 18, 1974	Millstone (BWR)	Fuel bundle transfer from fuel prep machine to spent fuel rack.	Unchanneled fuel assembly fell ~ 5 ft to pool floor during transfer from shipping container to inspection station.	Area evacuated - no radioactivity released. Bundle bowed while leaning against 2 spent fuel racks. Gas bubble escape was reported, but no radioiodine was detected. No damage to fuel pool. No evidence of fuel rod failure.	Incomplete closure of J-Hook on grapple when bundle bail not fully inserted.	Fuel grapple modified.	50245 421
9.	November 16, 1973	Zion (PWR)	Fuel loading into core.	Fuel bundle tipped against core baffle plate, moving 24-30 in.	Lower spacer grid cells distorted - No apparent damage to fuel rods.	Improper seating of bundle on lower core plate.	Alert fuel handling personnel; review fuel loading procedures.	--

(a) Based on incident reports assembled by W. J. Bailey and C. L. Wilson, Battelle Pacific Northwest Laboratories, Richland, WA.

or release of radioactive gases were observed, even for bundles dropped several feet through pool water, impacting on the bottom of the pool or on other bundles. Gaseous radiation releases register on monitors above the pool water when cladding is breached in the pool. Dropped fuel bundles generally are inspected by underwater viewing equipment to look for damage on visible rods and on fixtures.

In summary, normal fuel handling procedures within the pools appear to be satisfactory to preserve the mechanical integrity of irradiated Zircaloy- and stainless steel-clad fuel. In the few cases where fuel bundles were accidentally subjected to impacts, the incidence of breached cladding has been very low (two cases in nine dropped bundles and several thousand fuel handling sorties). Damage during shipping is less clearly defined, but has not been sufficient to cause handling problems in the receiving pool, nor even to be detectable in routine handling operations.

Mechanical Damage to the Pool

Mechanical damage to the pool liner is a possible consequence of fuel bundle and cask handling operations, which could influence fuel integrity if the pool water level drops, exposing the fuel to air. A case of liner damage occurred at the GE Morris pool in 1972, when an IF-100 shipping cask tipped against the loading pit wall, puncturing the stainless steel liner.(19) The damage area was crescent-shaped, approximately one foot long with a maximum parting of ∿1 1/2 inches. Pool water entered the space between the stainless steel liner and the concrete wall. A temporary repair was installed 27 hours after the incident, and a permanent repair was completed in 12 days, including design and fabrication of a 50-foot long access chamber.

One method to minimize damage to the pool in a cask drop involves a cask impact pad to absorb the energy of a dropped cask.(21) Recent fuel pool designs incorporate a cradle to prevent casks from tipping. Also the regulatory guides require either measures to prevent dropping of heavy loads or pool construction to withstand cask drops without causing leakage which could expose the fuel to air.

RANGE OF POOL STORAGE CONDITIONS

Currently the most valuable basis to define the long-term storage characteristics of spent nuclear fuel lies in an assessment of the behavior of the fuel inventory now in storage. The survey previously summarized in this report provides an initial assessment of that behavior. The large majority of fuel rods (>99%) successfully survived the relatively severe reactor environments, indicating excellent corrosion resistance.

Also potentially valuable are literature data on the behavior of materials exposed to conditions similar to those in the fuel pools. This section summarizes the range of fuel storage conditions defined in the survey, to provide a basis for comparison with published corrosion properties of similar materials. Figure 4 provides a perspective of a fuel pool configuration.

Spent fuel is stored in water pools at the following installations:

- Boiling Water Reactors (BWRs)
- Pressurized Water Reactors (PWRs)
- Independent Spent Fuel Storage Installations (ISFSIs)
- Research and Development Facilities
- ERDA Production Plants

The range of storage conditions was defined for a cross section of the above installations, including the following aspects: water volumes and temperatures, water chemistries, radioactivity concentrations, and the types of metals in contact with fuel bundle materials. Tables 5 (Canadian sites) and 6 (U.S. sites) compare information from ten sites, including the following:

- Three U.S. ISFSI pools
  - Allied General Nuclear Services, Barnwell, South Carolina(a)
  - General Electric Co., Morris Operation, Morris, IL
  - Nuclear Fuel Services (NFS), West Valley, NY
- Two U.S. nuclear power stations:
  - Three Mile Island Station, PWR, Middleton, PA
  - Dresden Station, BWR, Morris, IL

(a) Not yet licensed to receive spent fuel.

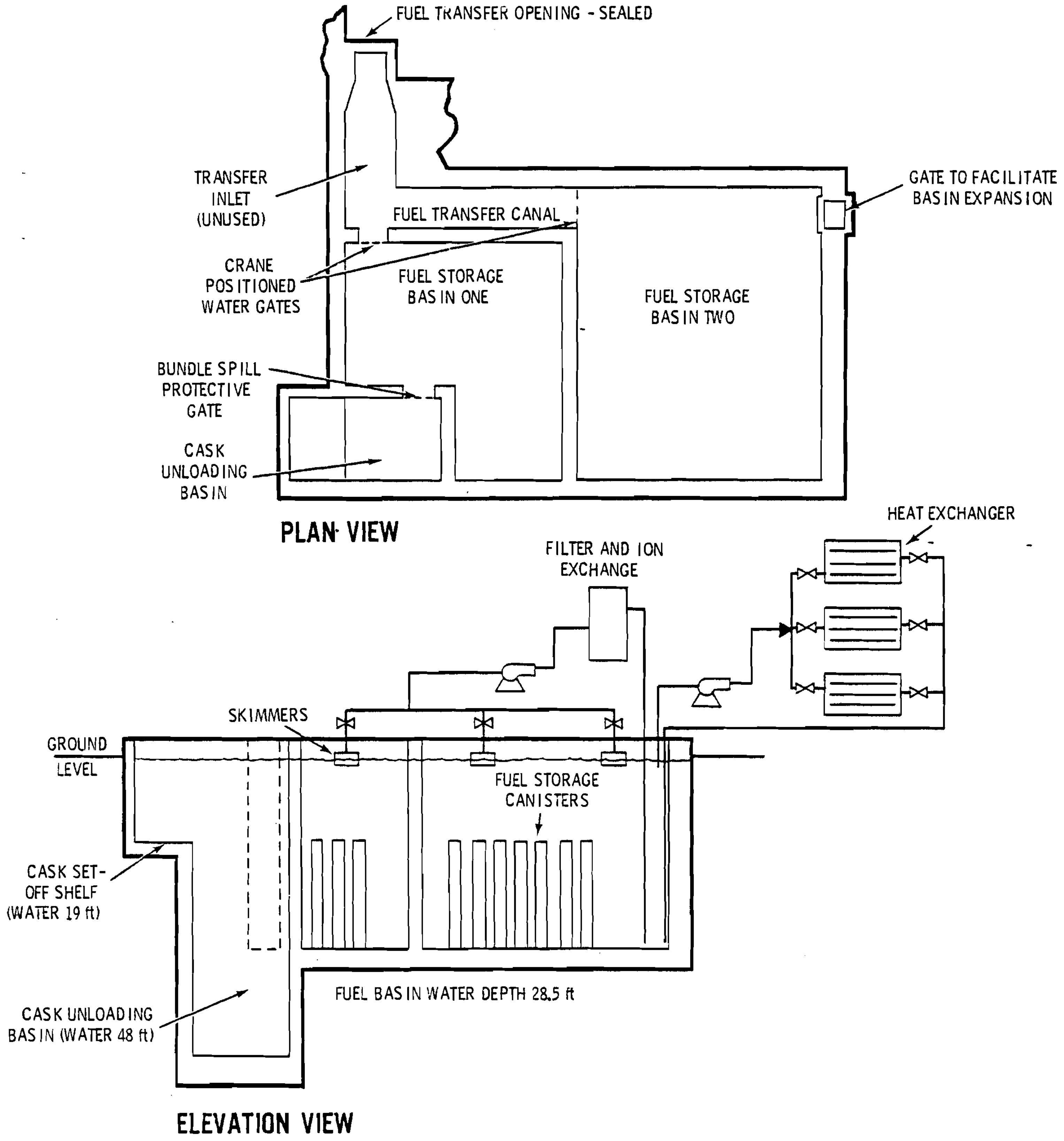

FIGURE 4. Schematic - Morris Operation Spent Fuel Storage Pool Facilities

- Four Canadian power stations (Pressurized Heavy Water Reactors, PHWs)
  - Bruce, Tiverton, Ontario
  - Douglas Point, Tiverton, Ontario
  - NPD, Rolphton, Ontario
  - Pickering, Toronto, Ontario
- One research and development facility:
  - Atomic Energy of Canada, Chalk River, Ontario, Canada

FUEL POOL WATER CHEMISTRIES

Tables 6 and 7 compare water chemistries for fuel storage pools at the 12 representative sites. Tables 8 and 9 compare water chemistry specifications for several pools. Pool chemistries are similar for pools at BWRs, ISFSIs, and most research and development facilities. The chemistries comprise oxygen-saturated deionized water. Some attempts have been made to reduce oxygen concentrations with hydrazine, without success due to the low reaction rates at pool temperatures except in radiation fields near the fuel bundles. However, hydrazine is added to some pools to convert iodine to water-soluble species, thereby reducing the concentration of air-borne iodine. The pH ranges of the deionized water basins are slightly acid to mildly basic (6.0 to 8.5). Some pools allow pH to reach the natural equilibrium with CO_2 ($\sim$5.8); at least one pool (NFS) uses caustic additions to control pH in the range of 6 to 8.

Major deviations from the water chemistry cited above occur at PWR pools. Boric acid is used as a chemical shim and lithium hydroxide as a pH control agent in PWR primary coolants. Boric acid also is added to PWR spent fuel storage pools. The PWR primary coolant and pool water mix during fuel discharges and refuelling, so it is easier to maintain balanced chemistries if both systems have similar compositions. Boric acid has similar concentrations in PWR pool and primary systems; lithium frequently is added to the primary system, but not to the pool water. The pH's of PWR pools are in the acidic range (4.5 to 5.5), even though some PWR pool specifications permit basic pool chemistries (Table 8).

TABLE 6. Characteristics of Canadian Fuel Storage Pools

	AECL-Chalk River		Ontario Hydro					
	NRU	NRX	Bruce A		Douglas Point	NPD	Pickering A	
Water Volume, gal	237,000	174,000	2,490,000 (2 pools)		350,000	29,200	1,120,000 (2 pools)	
Pool Dimensions:	several pools	5 pools						
length, ft			136	150	68.4	18.5	110	112
width, ft			31.8	60	25	10	54	56
depth - fuel transfer, ft		10-14	14		12.9		17.5	
fuel storage, ft	29	20	20	27.5	23.9	16	27.5	26.5
Water Flow Rate to Heat Exchanger, gpm	25		4200 max	1800	400	90	5200 max	1000
Pool Temperature Range, °C	18	18	21-29		26-32	32-40	32	30
Water Chemistry								
pH range	6.5-9.5	6.5-9.5	5.5-8.0				5.5-6.5	
Cl^-, ppm			<0.3				<0.5	
Conductivity, μmho/cm	3.7	3.7	2-5				<5	
Heavy Metals (Fe, Cu, Hg, Ni), ppm								
Radioactivity Control - Pool Water								
Dissolved	Ion Exchange	Ion Exchange	Ion Exchange		Ion Exchange	Ion Exchange	Ion Exchange	
Particulate	Filtration	Filtration	Filtration		Filtration	Filtration	Filtration	
Radioactivity Concentrations*								
Total μCi/mℓ	9×10^{-4}							
Iodine "							$<10^{-1}$	
Cesium " (^{134}Cs, ^{137}Cs)								
Cobalt "								
Pool Storage Materials								
Grapples	SS	SS	Al + SS		304 SS	Al + SS	304 SS	
Canisters	Al or SS	Al or SS	304 SS		304 SS	304 SS	304 SS	
Storage Racks	SS	SS			430 SS			
Pool Structure	Concrete	Concrete + Epoxy	Concrete + Epoxy		Concrete	Concrete	Concrete(a)	
Pool Liner	Tile	Monel and SS	304 SS (partial)		304 SS	304 SS	Epoxy, Fiberglass	
Pool Building	Stl frame + Brick	Stl + woodframe	Concrete + St		Concrete + St	Concrete	Concrete + Stl. Steel	
Heat Exchanger	Cu Alloy	Cu Alloy	304L SS					
Pool Storage Capacity								
Assemblies (1977)	n.a.	~1400	250,000		85,800	2,800	250,000	
Current Inventory, Metric Tons**	2.65	17.1	0		116	2.6	1095	
First Fuel Storage	1957	1947	1977		1968	1964	1971	

(a) One concrete tank inside another concrete tank, for double containment and leak collection.

SS = Stainless Steel
n.a. = not available

* all others - no specs
**metric tons of U as of January 1, 1977

TABLE 7. Characteristics of U.S. Fuel Storage Pools

	AGNS[a]	GE Morris	NFS	Dresden BWR	Three-Mile Island PWR
Water Volume, gal	1.3 x 10^6 (4 pools)	675,000	830,000	1.04 x 10^6 (3 pools)	260,000
Pool Dimensions:					
length, ft			75		210
width, ft			29		30
depth - fuel transfer, ft		48	44		
fuel storage, ft		29	29	38	30
Water Flow Rate to Heat Exchanger, gpm	2,000	750	2400	700 x 2 per pool	
Pool Temperature Range, °C	(a)	25-35 norm 41 max	27-32 41 max	27-32 norm	
Water Chemistry					
pH range	6-8	4.4-9 (5.8 mean)	6-8	6.5-8.5	5.2-5.5
Cl^-, ppm		~0.02 (norm)		<0.1	0.01
Conductivity, μmho/cm		~1	20	0.5-1	8.15-8.50
Boron, ppm		~1			2120-2140
Lithium, ppm					low (n.a.)
Heavy Metals (Fe, Cu, Hg, Ni), ppm				<0.1	n.a.
Radioactivity Control - Pool Water			(100 gpm)	Ion Exchange	
Dissolved	Ion Exchange	Ion Exchange	Ion Exchange	Precoat	Ion Exchange
Particulate	Etched Disk Filter 300 gpm	Precoat Filter 250 gpm	Diatom Earth 500 gpm	Filter	Cart. & Precoat
Radioactivity Concentrations					
Total μCi/mℓ	5 x 10^{-2} (max)[a]	4 x 10^{-4} (norm)	8 x 10^{-4} (norm)		5 x 10^{-2}[b]
Iodine "		--			
Cesium " (^{134}Cs, ^{137}Cs)		2.8 x 10^{-4}			4 x 10^{-3}
Cobalt "		0.8 x 10^{-4}			2 x 10^{-2}[b]
Pool Storage Materials					
Grapples	304 SS	SS	SS	Cr-plated steel	SS
Canisters	5083 Al	SS	Al	Al & SS	
Storage Racks	6061 Al	SS	Al	Al & SS	
Pool Structure	Concrete	Concrete	Concrete	Concrete	Concrete
Pool Liner	304 SS	SS	Paint + SS	SS	SS
Pool Building	Concrete	Concrete	Steel	Concrete	Concrete
Heat Exchanger	316 SS	Finned SS	SS		
Pool Storage Capacity					
Metric Tons	360	750	280		
Equivalent BWR Assemblies		3,735		3,184	
Equivalent PWR Assemblies		1,560			~300
Current Inventory, Metric Tons	0	225 (Mar. 1, 1977)	162		
First Fuel Storage		1972	1965	1960	1976
Fuel Storage Rack Spacings					
Original, inches			20	10.0	21.0
Stainless Steel, inches				7.3	13.0
Stainless Steel + Boron, inches				6.5	10.5

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[a] Has not begun to receive fuel (1977).

[b] Maximum during refuelling.

n.a. = not analyzed

TABLE 8. Spent Fuel Pool Water Quality Specifications(a)

	PWR	
	Normal	Transient Limits
pH (77°F)	4.5 - 6.0(b)	--
Conductivity @25°C (μmho/cm)	1 - 30	--
Chloride ion (ppm, max)	0.15	1.5
Fluoride ion (ppm, max)	0.1	1.5
Oxygen	Saturated	Saturated
Total suspended solids (ppm, max)	1.0	--
Temperature (°C)	50	100
Boric acid (as ppm-B(c))	0 - 2000 1950 - 2250	--
Activity level (μCi/mℓ)	0.05 varies with tech spec.	
Solid filtration (microns)	1 - 5	--

(a) May vary somewhat from pool to pool.

(b) Some fuel pool pH range specifications are 4.5 - 8.0; a few pools have a range of 4.0 - 10.6.

(c) Some PWR fuel pool specifications also indicate a lithium concentration range of 0.2 - 2.0 ppm, corresponding to values for PWR primary systems.

The boric acid presents some problems regarding water purity control by ion exchange. Cleanup trains consisting of filters and boron-loaded ion exchange resins are used at PWR pools.

Particulate and dissolved impurities are controlled by combinations of ion exchange and filters. Filter types are indicated in Table 7. Skimmers remove materials which float on the water surface to maintain visual clarity and reduce radioactivity. Storage pool cleanup systems are discussed in more detail elsewhere.(22)

Table 9 provides water chemistry specifications for U.S. BWR, Bruce (Canadian), and GE-Morris pools, all using deionized water.

TABLE 9. Fuel Pool Water Chemistry Specifications

	BWR	Bruce Station Ontario Hydro			GE-Morris
		Optimum	Action Limits	Unacceptable Range	
Conductivity, μmho/cm, 25°C	≤ 3*	2	5	10	--
Chloride (as Cl^-), ppm	≤ 0.5	<0.3	<0.5	>1	10
pH	5.3-7.5	5.5-8.0	<5, >8	<4, >9	4.5-9.0
Heavy Elements, ppm	<0.1	--	--	--	--
Total Insolubles, ppm	<1	--	--	--	--
Radioactivity μCi/ml, max					0.02(a)

(a) Transient limit, 0.1 μCi/ml

* ≤ 1.5 at some pools

The usual sampling frequencies for pool water chemistry are weekly or twice per week. Conductivity, chloride, and pH are the analyses commonly performed. Boron and sometimes lithium are analyzed for PWR pools. Analyses seldom include heavy metals. When analyzed, heavy metal concentrations were low (<0.1 ppm). To illustrate, an analysis of a 1-gm residue from evaporation of 12 ℓ of GE-Morris pool water indicated that concentrations of all metallic species from calcium through uranium were individually 0.004 ppm or below, based on data provided by Dr. E. E. Voiland, Manager of the G. E. Morris facility.

Basin chemistry control generally does not present operational problems. However, at one time, $NaNO_3$ was used as an antifreeze agent in fuel shipping casks, causing contamination of fuel pools. As an example, nitrate concentrations at the G. E. Morris pool reached ∿200 ppm. The nitrate caused rapid depletion of the Powdex ion exchanger and relatively high conductivities (∿250 μmho/cm, compared to current values of ∿1). After nitrate use was discontinued, the NO_3^- concentrations gradually decreased from 200 ppm in January 1972 to less than 0.6 ppm in January 1977. Chloride remained at ∿3 ppm until NO_3^- dropped to ∿3 ppm, then Cl^- decreased to less than 0.02 ppm.

Some older concrete-lined pools at R&D facilities operate with relatively high impurity levels in pool waters. In R&D facility pools which use chloride for microbiological control, chloride concentrations have reached several hundred ppm. In the newer pools operating with high-purity water, biological growth is not frequently a problem, based on this survey and observations reported elsewhere.(22) One pool (ECF) uses refrigeration to maintain low temperatures to suppress algae growth.

Effects of Boric Acid Pool Chemistry

Boric acid has functioned successfully as a reactivity control agent in PWR primary systems. It has proved to be compatible with fuel bundle and primary system materials at reactor operating temperatures. Boric acid chemistry also is used in PWR fuel pools, as indicated earlier. The pH range is ∿4.5 to 5.5 at pool temperatures. The pool water also contains some lithium, from mixing with the reactor coolant, but the concentrations generally are not controlled by lithium additions to the pool water. Lithium concentrations frequently are below the range maintained in the PWR primary systems (0.2 to 2 ppm). The major differences from PWR primary system operation are the lower temperature regime and the difference in oxygen concentration. During PWR operation, the oxygen concentrations are maintained at less than 0.01 ppm by hydrazine additions and hydrogen in the coolant. In the fuel pool water, oxygen is presumed to be at saturation in equilibrium with the atmosphere (5 to 8 ppm). As indicated earlier, hydrazine additions are not effective to maintain low oxygen concentrations in the large volume of pool water.

The boric acid has presented some problems in regard to ion exchange operation to maintain water purity. However, ion exchange resins have been developed which continue to absorb species such as chloride ion while heavily loaded with boric acid.

In most cases, pool and fuel bundle materials have appeared to function satisfactorily in boric acid fuel pool chemistry, but very few detailed analyses of the materials are available. Aluminum corrosion behavior has been substandard in some borated pools. One utility had 6061 aluminum canisters residing directly on the stainless steel liner at ∿110°F. Substantial corrosion developed on the aluminum canisters, forming aluminum borate ($Al_4B_2O_9$) which flaked off. Considerable aluminum corrosion also occurred in crevices on the canisters, perhaps due to oxygen concentration cells. Insulators between the canisters and the liner minimize galvanic corrosion of the aluminum. Cypress wood and teflon were first used as insulators. However, both were deficient. The cypress deteriorated in water, and the teflon did not meet the criterion of resistance to 10^8 rad of radiation exposure. The cypress insulators are being replaced with alumina and the teflon is being replaced with silicone resin.

FUEL POOL AND FUEL ROD TEMPERATURES

Heat generated in spent fuel by radioactive decay is dissipated in the fuel pools, elevating the water temperatures. The decay heat at three days after discharge is about 0.3% of the full power heat release. After five years the value is only ∿0.06% of the full power value.(23)

Reactor pool specifications generally call for fuel pool temperatures of $\leq$ 120°F (40°C) with full heat exchange capacity; $\leq$150°F (66°C) with one heat exchanger and aged fuel and 212°F (100°C) with freshly discharged fuel and loss of heat exchange capacity. However, there are no reports that pool temperatures have approached 100°C. The maximum actual pool temperature reported in the survey was 49°C.

A knowledge of the temperature at the fuel rod surface is important in defining rates of corrosion and other chemical interactions. No direct measurements appear to have been made on fuel rod surfaces, but calculated fuel rod surface temperatures supplied to J. Tanaka of Ontario Hydro indicate the following:

Fresh, High-Exposure Fuel (45-day cooling, 32°C bulk water): $\Delta T = 10°C$
Aged, High-Exposure Fuel (4-year cooling, 32°C bulk water): $\Delta T = 1°C$

Independent calculations made by C. W. Stewart at Battelle Pacific Northwest Laboratories indicate values in the same range as those reported above for surface temperatures on pool-stored fuel.

FUEL ROD RADIATION LEVELS

Radiation contributes to the environment on both sides of the spent fuel rod surfaces. Both beta and gamma emitters occur on both sides of the cladding. However, the large neutron component which was present in the reactor is missing, and the gamma component has dropped by one to two orders of magnitude, to $>10^6$ R/hr immediately after reactor shutdown on the most radioactive surfaces.(24) The radiation decay will roughly parallel heat decay. Estimates provided by Ontario Hydro indicate the following values for radiation levels on their fuel in pool storage:

Fresh, High-Exposure Fuel - Equiv. to 10^5 R/hr at 1 ft in air
Aged (5 y), High-Exposure Fuel - Equiv. to 3×10^2 R/hr at 1 ft in air

FUEL POOL MATERIALS

The fuel pool materials identified in the survey are summarized in Table 10. After irradiation, the fuel bundles are contacted with steel grapples, used to remove them from the reactor core. The bundles are transferred to stainless steel viewing trays where they are inspected. They are then transferred to storage canisters (Figure 2). The canister types used at the GE-Morris pool are shown in Figures 5 and 6. They are constructed of alloy 304 stainless steel pipes with a solid steel plate welded across the bottom. The pipes are welded together into modules of either nine 8-inch

TABLE 10. Summary of Materials in Fuel Pools

Component	Sub-Components	Material[a]	Alloy
Wall	--	Reinforced Concrete	--
Pool Liner:	--	Stainless Steel Epoxy, Fiberglass	304
Heat Exchanger[b]	--	Stainless Steel	304/316
Filter:	Vessel Filter Elements:	Stainless Steel Stainless Steel Diatom. Earth, Fiber	304L 304L
Recirculating Pumps:	Casing, Shaft Impeller	Stainless Steel Bronze	316
Demin. Water Return Pump	Casing, Impeller, Shaft	Stainless Steel	316
Deionization Unit	Tank	Stainless Steel	304L
Cask Head Support Racks	--	Stainless Steel	304L
Gates and Guides	--	Stainless Steel	304L
Canister Storage Racks:	Racks:	Stainless Steel Aluminum	304L 6061-T6
	Embedded Supports	Stainless Steel	304L
Fuel Storage Canisters:[c]	--	Stainless Steel Aluminum	304L 5083/5086/6063
Leaker Can Support Racks:	--	Stainless Steel Carbon Steel, Epoxy-Coated	-- --
Control Rod Cluster Storage Racks	--	Stainless Steel	--
Portable Offgas Hoods:	--	Stainless Steel Aluminum	--
Cask Handling Crane	Cable & Grapple	Stainless Steel	--
Canister Crane	Cable & Grapple	Stainless Steel	--
Fuel Transfer Conveyer	--	Stainless Steel	304L
Insulators	--	Tygon, Neoprene	
Cask Impact Pad:	Cladding Pad (not exposed to H_2O)	Stainless Steel Carbon Steel (3-1/2-in. thick)	
	Honeycomb (not exposed to H_2O)	Aluminum	

(a) Types identified in survey; other types may be used in some pools.

(b) Carbon steel tubes were originally installed in heat exchangers at one pool; severe rusting caused a visibility problem in the pool water, resulting in retubing with stainless steel. Copper alloy tubes are used at one R&D facility pool.

(c) Some canister walls contain boron-impregnated aluminum for reactivity control, clad with stainless steel or aluminum.

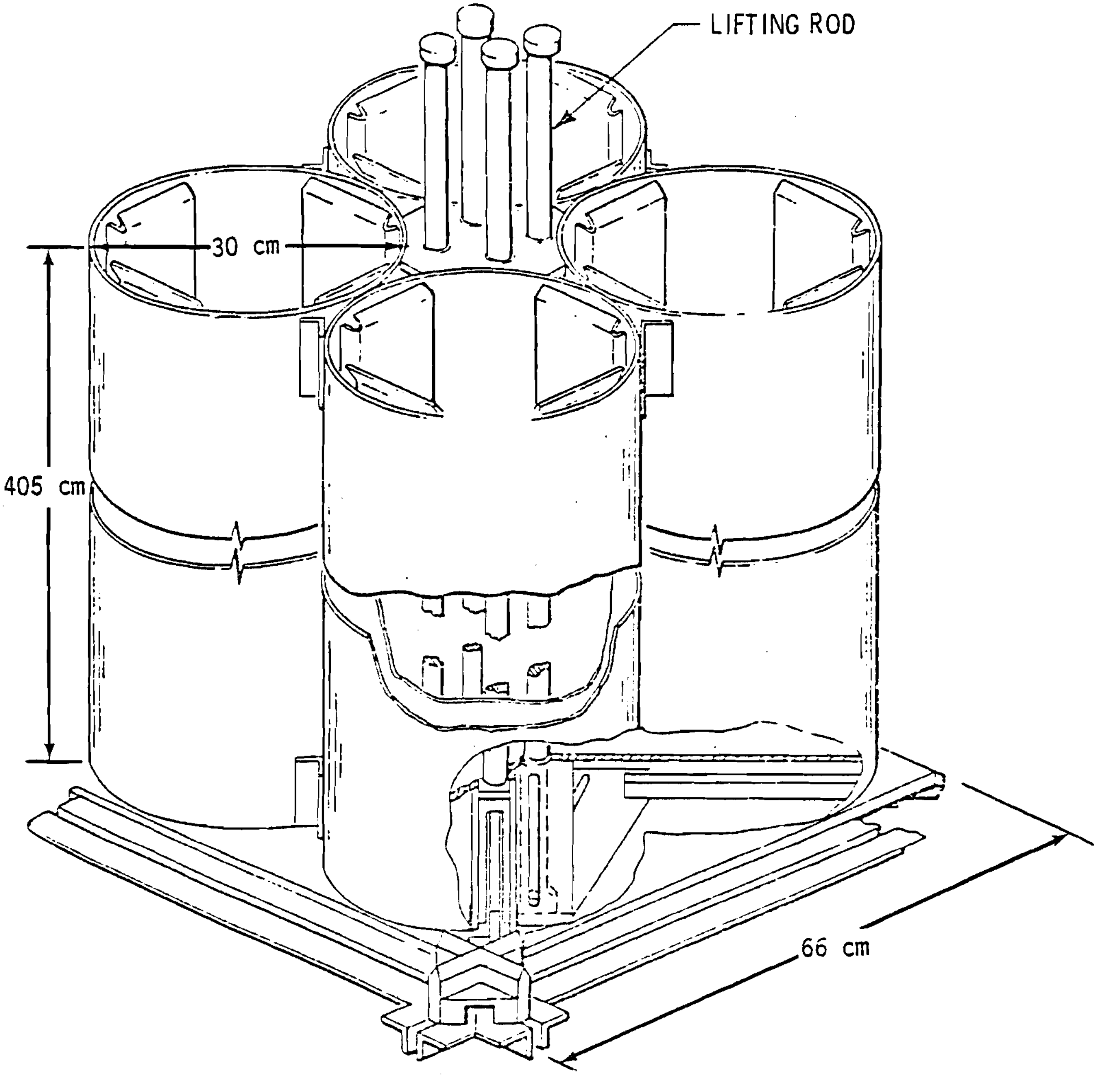

FIGURE 5. PWR Stainless Steel Fuel Storage Module - G.E. Morris Operation.(25)

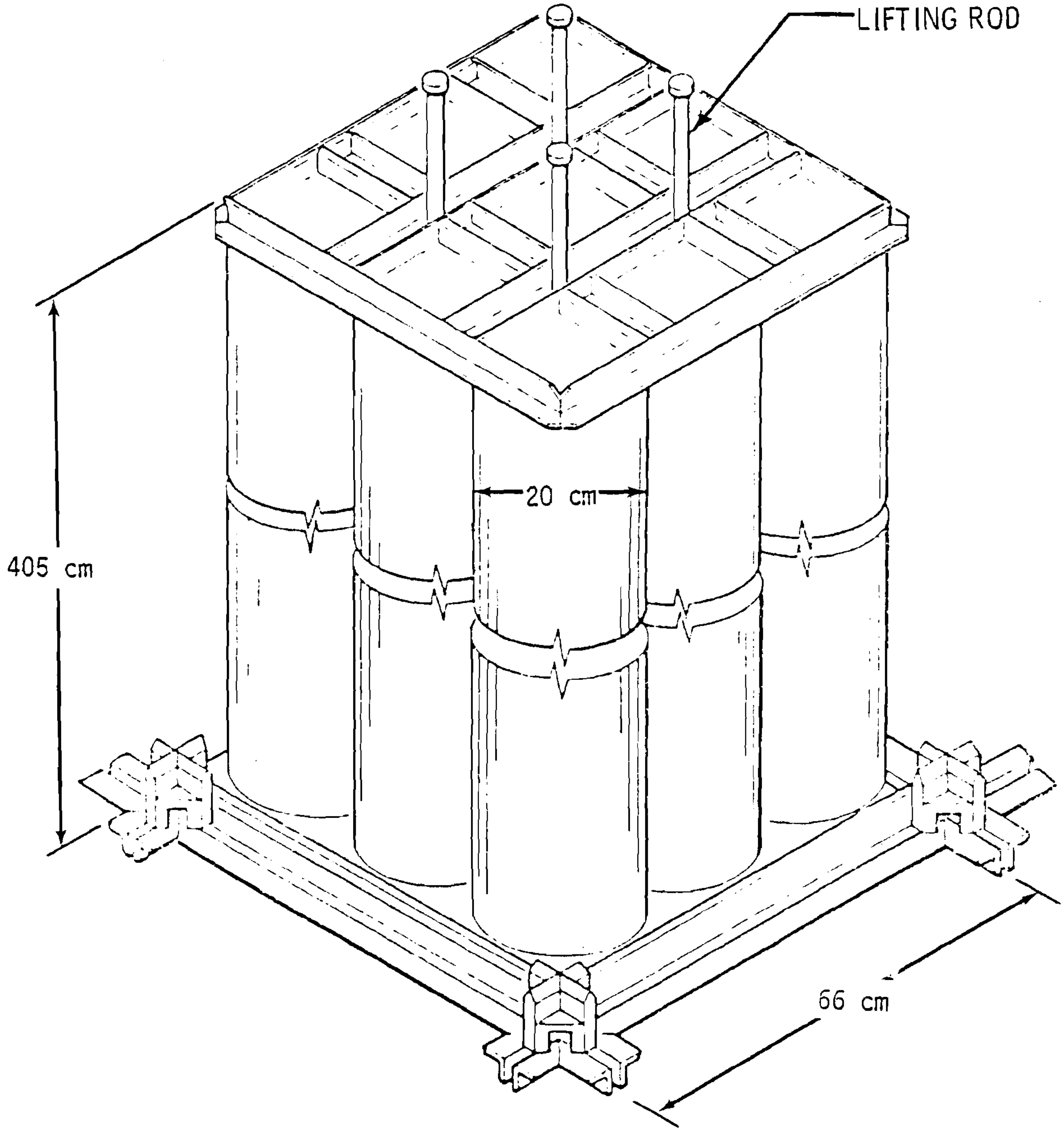

FIGURE 6. BWR Stainless Steel Fuel Storage Module - G.E. Morris Operation.(25)

schedule 10 pipes for BWR bundle storage or four 12-inch schedule 5 pipes for PWR bundle storage. The modules have locking devices which fix them to stainless steel racks mounted to the bottom of the pool. Both modules fit the same rack position. During pool residence, the only materials likely to contact the stored fuel would be the stainless steel canisters and trace impurities transported in the water.

Some fuel canister designs have square cross sections (Figure 7). The materials may be either stainless steel or aluminum alloys. In reactor pools, the bottom of the canister generally has a screen and sometimes a nozzle for water circulation to promote cooling of the fuel bundles. Some pool canisters are seated on racks mounted to the bottom of the pool; in some pools the canisters reside on the bottom of the pool.

The contacts between the fuel bundles and the canisters will occur where the lower nozzle or tie plate rests on the plate or screen on the bottom of the canister; also, at locations where upper parts of the bundle contact the edges of the canister or guides and spacers installed inside the canister. The most probable contacts would be between the canister and the tieplates, but some contacts between the canister walls and the spacer grids or fuel rods are possible, particularly if there is some bundle distortion.

Other fuel pool materials which do not generally come in contact with the fuel bundles include the stainless steel pool liner (painted concrete walls and fiberglass are used in some pools); carbon steel and stainless steel piping; stainless steel and Inconel heat exchangers; ion exchange resins; various filter materials, including diatomaceous earth and fiber types.

Fuel bundles are contained inside stainless steel fuel baskets during shipping. Boron carbide clad in stainless steel provides criticality control in the fuel baskets.

Storage configurations in Ontario Hydro pools involve placement of 10 to 32 bundles on stainless steel trays in close-packed arrays. The bundles may be oriented either vertically or horizontally, depending on the reactor site.

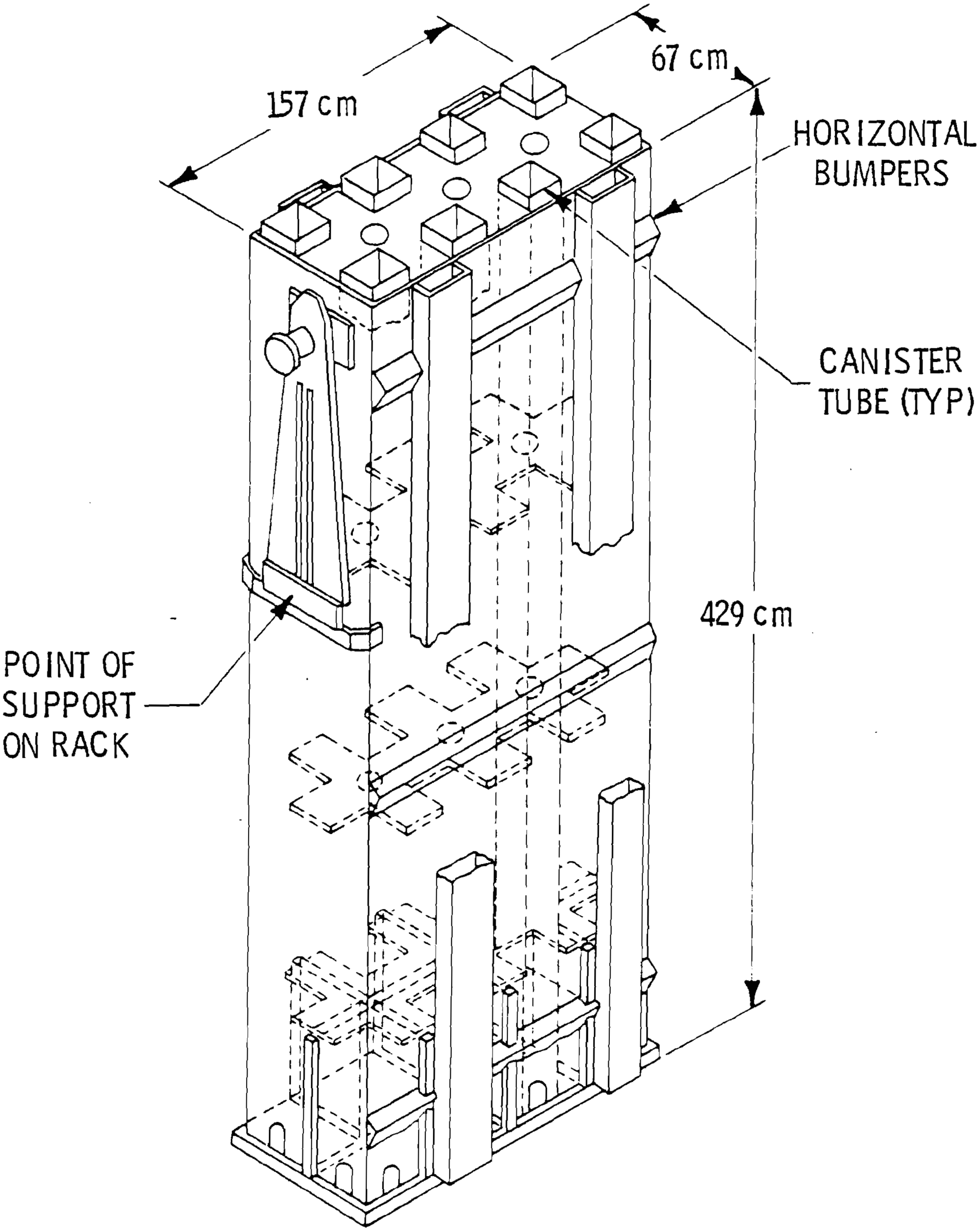

FIGURE 7. Fuel Storage Canister(3)

FUEL BUNDLE MATERIALS

The range of materials interactions between the fuel bundle and materials in the storage pool are potentially important to define possible degradation mechanisms. Equally important is the range of materials interactions within the fuel bundle. Figure 8 shows a diagram of a PWR fuel rod. Figures 9 and 10 show diagrams of BWR and PWR fuel bundles, respectively. Table 11 summarizes the range of materials in stainless- and Zircaloy-clad fuel bundles.

MATRIX OF FUEL STORAGE CONDITIONS

Table 12 shows the range of interactions of fuel bundle materials, canisters and fuel pool water chemistries. For example, the combination of Zircaloy fuel cladding, stored in stainless steel canisters in deionized water (B-a-2) represents one condition at the GE-Morris plant, the other being A-a-2. All eight combinations have been identified in one or more pools.

In a mechanistic assessment of long-term effects of pool storage on fuel bundle materials, possible effects of the above combinations need to be considered. At the present level of perception, stored fuel appears to behave satisfactorily in all of the combinations covered by the survey.

GALVANIC COUPLES

From the foregoing discussion the following ranges of galvanic couples exist within fuel bundles and between bundles and fuel storage canisters:

Within bundles:

Zircaloy - Inconel
Zircaloy - Stainless Steel
Inconel - Stainless Steel

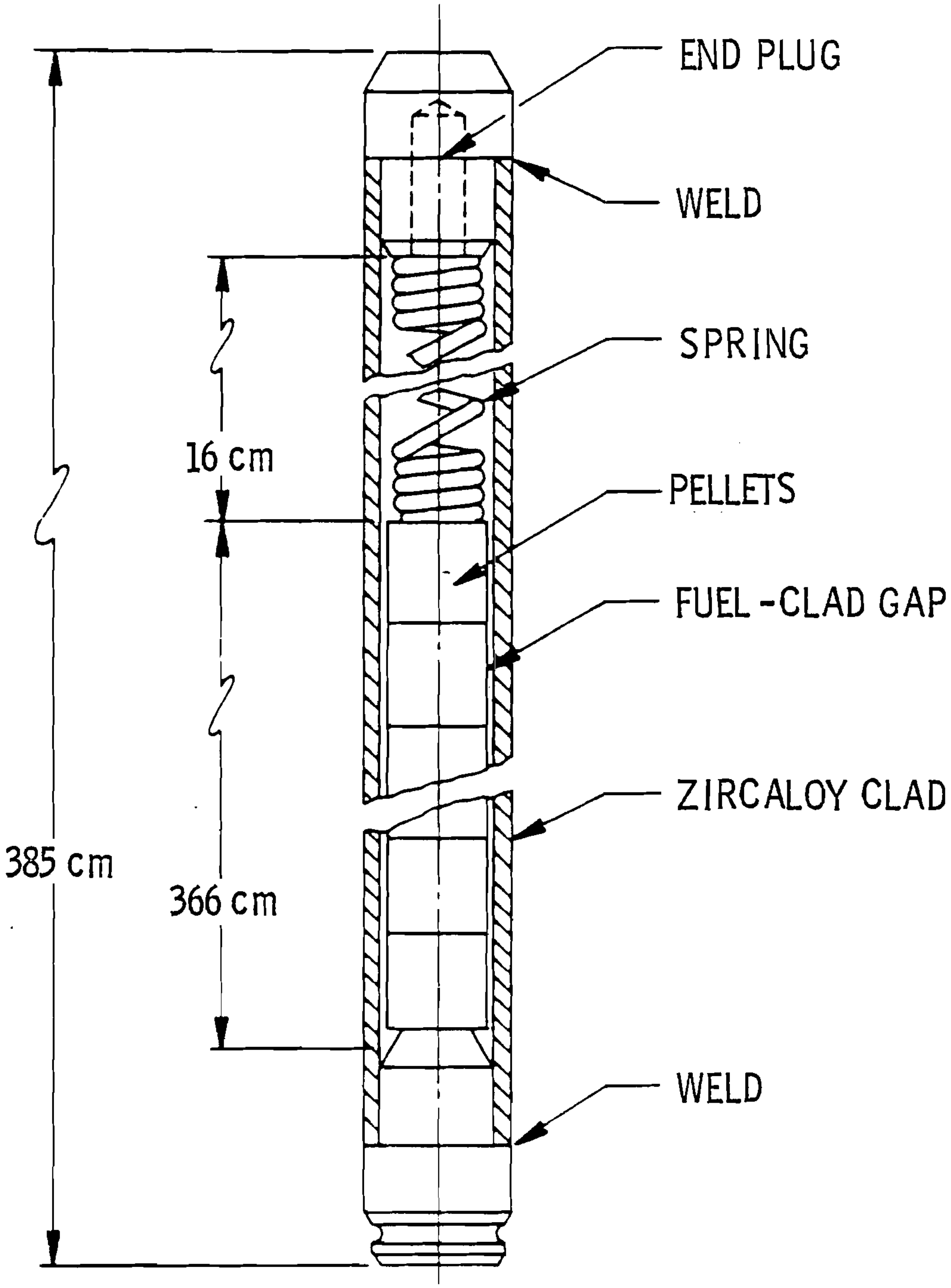

FIGURE 8. Typical PWR Fuel Rod(3)

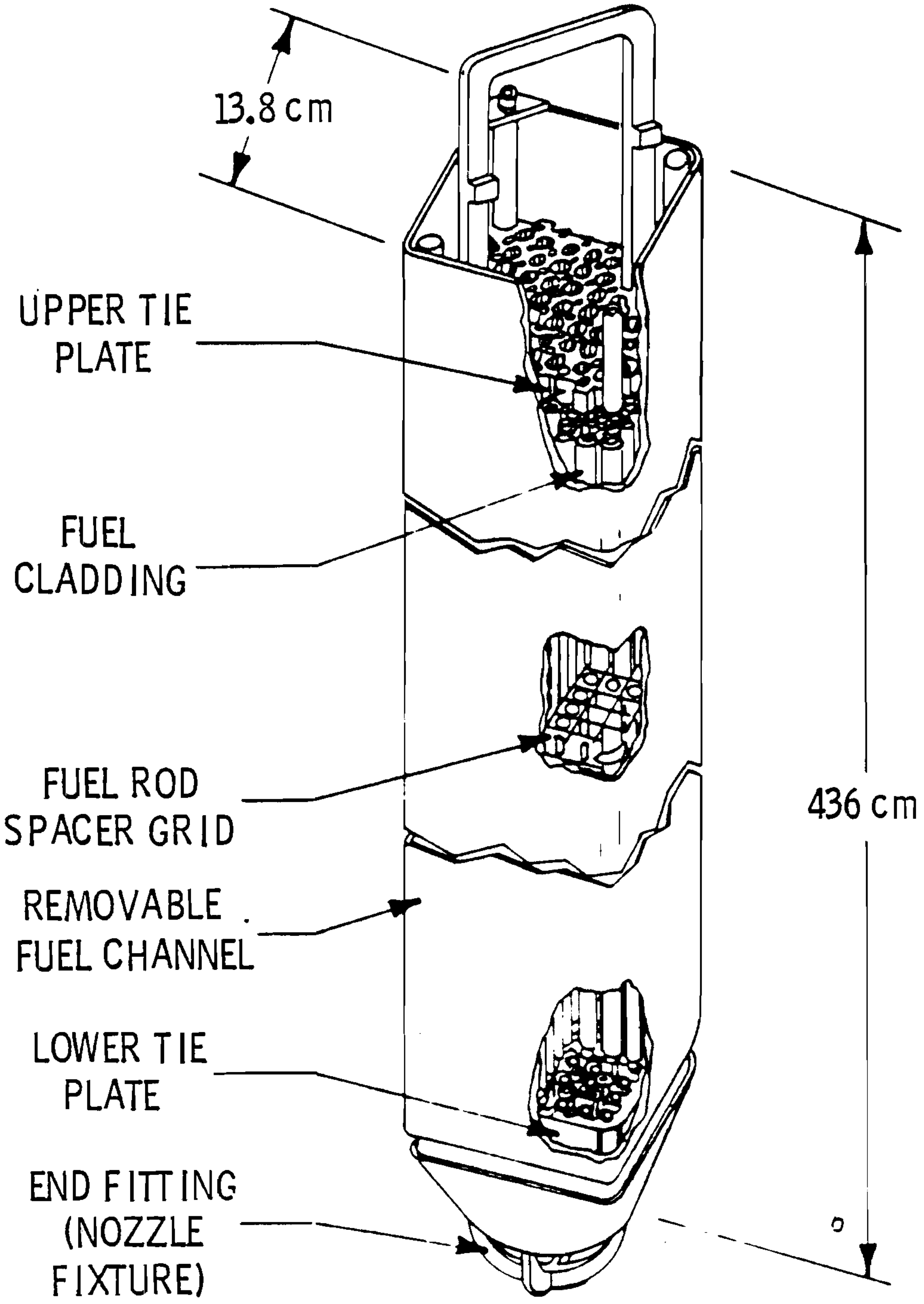

FIGURE 9. BWR Fuel Assembly(3)

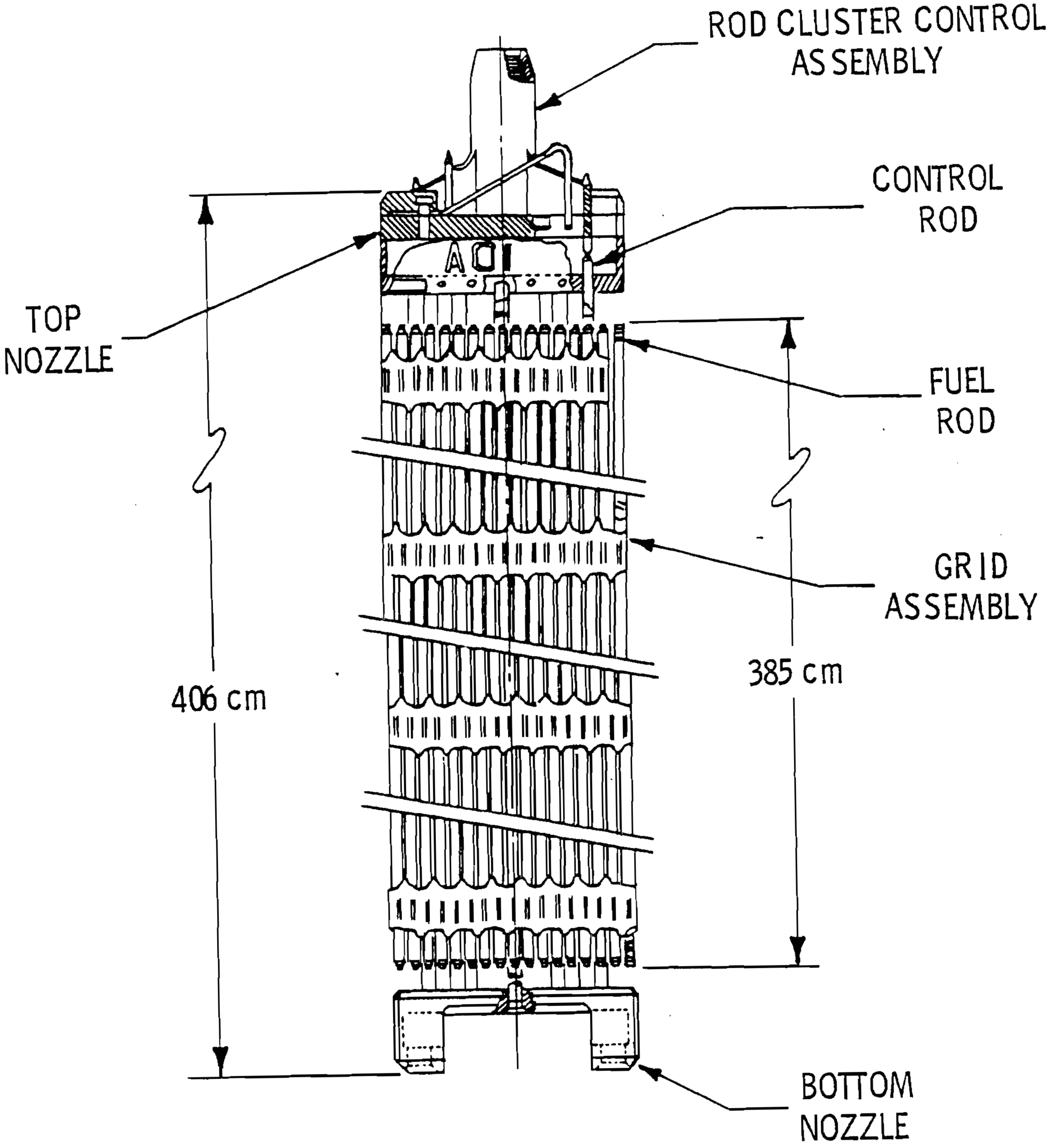

FIGURE 10. PWR Fuel Assembly(3)

TABLE 11. Fuel Bundle Materials[a]

Component	Sub-Components	Material	Alloy
Fuel Rods:	--	Zircaloy:[b]	Zry-2 (BWR) Zry-4 (PWR)
		Stainless Steel:	304 348H
Fuel Spacers:	Grid:	Stainless Steel	304
		Inconel	718
		Zircaloy	Zry-4
	Springs:	Inconel	718
Upper Tie Plates:	Bail/Tie Plate	Stainless Steel	304
	Bolts/Nuts	Stainless Steel	304
		Inconel	600
	Springs	Inconel	718
Lower Tie Plates:	Tie Plate/Nozzle	Stainless Steel	304
Tie Rods:		Zircaloy Stainless Steel	--

---

[a] Materials identified in the survey. Some variations on material and alloy specifications may occur.

[b] Both Zry-2 and Zry-4 are used for cladding in Canadian reactors. Zircaloy is the only material in Canadian fuel bundles, except for small amounts of beryllium used to braze the appendages.

Between Bundles and Canisters:

Stainless Steel - Stainless Steel
Stainless Steel - Aluminum Alloys

In some cases, Zircaloy-stainless steel and Zircaloy-aluminum couples may exist in canister-stored fuel, in bundles with bowed fuel rods. However, the general case will be that the Zircaloy fuel bundle components will form galvanic couples with the canisters only by way of stainless steel nozzles and/or tie plates, rather than by direct canister/Zircaloy contacts.

The implications of galvanic couples to fuel bundle degradation are discussed in a later section.

TABLE 12. Matrix of Fuel Storage Conditions

Fuel Cladding Material	Canister Material	Water Chemistry
A. Stainless Steel	a. Stainless Steel	1. Boric Acid
		2. Deionized
	b. Aluminum Alloy	1. Boric Acid
		2. Deionized
B. Zircaloy	a. Stainless Steel	1. Boric Acid
		2. Deionized Water
	b. Aluminum Alloy	1. Boric Acid
		2. Deionized Water

RADIOCHEMICAL CHARACTERIZATION OF FUEL POOL WATERS

The radioactive species in fuel pool waters are monitored periodically (normally once or twice per week) for beta and gamma activity, and some analyses are made for alpha activity. Rates of radioactivity buildup on filters and ion exchange resins also are monitored. Monitors for airborne radioactivity operate continuously above the pools.

The sources of radioactivity in pool waters are activation products and fission products. The sources of the activation products are crud deposits and corrosion films on the fuel bundle surfaces. The fission products arise from bundles with rods which failed in-reactor or from intact bundles which adsorbed circulating fission products. The principal activation products released to pool waters, their half lives and source reactions are given in Table 13. The principal fission products in pool waters appear in Table 14. Reference 3 gives inventories of fission products in fuel rods as a function of time after reactor discharge. Cesium, tritium, cerium, strontium and the iodines are the principal fission products in reactor pools. For aged fuel, the most significant isotopes from the standpoint of pool waste management are: ^{137}Cs, ^{3}H, ^{60}Co, ^{90}Sr, and ^{144}Ce Pr.(22) The principal isotopes and their concentration ranges are given in Table 15.

TABLE 13. Principal Activation Products Released from Fuel Bundles During Pool Storage(26)

Nuclide	Half-Life	Production Reaction
Co-60	5.3 y	Co-59(n-γ)
Co-58	72 d	Ni-58(n-p)
Cr-51	28 d	Cr-50(n-γ)
Fe-59	45 d	Fe-58(n-γ)
Mn-54	310 d	Fe-54(n-p)
Zn-65	243 d	Zn-64(n-γ)
Ni-65*	2.5 d	Ni-64(n-γ)
W-187*	24 h	W-186(n-γ)

---

*Only significant at reactor pools.

TABLE 14. Principal Fission Products Released to Fuel Pool Waters(22)

Isotope	Half-Life
^{131}I	8.05 d
^{134}Cs	2.1 y
^{137}Cs	30 y
^{3}H	12.3 y
^{90}Sr	28.8 y
^{144}Ce	285 d
^{106}Ru-^{106}Rh	1.0 y-2.2 hr
^{95}Zr-^{95}Nb	65 d-35 d

NOTE: Alpha activities also are monitored. At a German spent fuel pool(27), fission product activities were 5 x 10-5 μCi/cc; alpha activities were an order of magnitude lower.

At G. E. Morris, alpha activities in pool water are $<10^{-6}$ μCi/ml.

TABLE 15. Radionuclide Concentrations In Fuel Storage Pools(a) μci/ml(22)

Radionuclide*	Current Generation Basins With High-Integrity Fuel	Early Generation Basins	Older ERDA Basins
^{3}H		1×10^{-3}	
^{54}Mn		1×10^{-5}	
^{58}Co	5×10^{-6} - 3×10^{-5}	2×10^{-5} - 1×10^{-4}	
^{60}Co	5×10^{-6} - 1×10^{-4}	1×10^{-5} - 1×10^{-4}	5×10^{-5}
^{90}Sr		2×10^{-5}	2×10^{-2}
^{131}I	1×10^{-7}	1×10^{-6}	
^{134}Cs	1×10^{-5} - 1×10^{-4}	3×10^{-4}	3×10^{-3}
^{137}Cs	3×10^{-5} - 1×10^{-4}	5×10^{-4} - 1×10^{-3}	3×10^{-2}
^{140}Ba		1×10^{-5}	
^{144}CePr			3×10^{-3}
Pu			2×10^{-5}
Total, μCi/ml	1×10^{-7} - 1×10^{-3}	1×10^{-3} - 1×10^{-2}	1×10^{-2} - 5×10^{-1}
Dose Rate,(b) mrem/hr	<1	<5	5-50

(a) At equilibrium conditions; higher values are generally present immediately following reactor fuel discharge.

(b) Dose rates at reactor pools may approach 40-100 mrem/hr on occasion during cleanup system upsets and following fuel discharges.

* At some pools, the following isotopes also are significant: ^{141}Ce, ^{140}La, ^{63}Ni, ^{89}Sr, ^{95}Zr/^{95}Nb, ^{65}Zn.

The activation products come principally from corrosion products from the reactor primary circuits which deposit as crud layers on fuel bundle surfaces (Fe_2O_3, Fe_3O_4, spinels, sometimes oxides of nickel and copper). Crud layers may be 25 to 50 μm thick, particularly on BWR fuel rods. The layers generally are thinner and sometimes are almost absent on PWR fuel rods. The crud layers are quite insoluble at fuel pool temperatures, but do release some particulates to the pool water, together with traces of soluble species. The corrosion products on Zircaloy rods (ZrO_2) also have low solubilities and generally resist spallation.

There are substantial differences in the inventories of radioactive species at reactor pools and at other pool facilities. During fuel discharges, reactor pool contamination levels rise from a combination of dissolved and particulate species in the reactor primary coolant, which mixes with pool water, and from species released from fuel bundle surfaces. Short-lived species such as iodine isotopes appear in reactor pools, but rarely at other pools. Tritium also is substantially higher in reactor pools. Loose crud is released from the fuel bundle surfaces during handling in the reactor pool. Radiation levels at reactor pools often rise to $\sim 10^{-2}$ μCi/ml during discharges, but can be controlled at 10^{-3} to 10^{-4} μCi/ml after fuel discharges are completed. Radiation levels at reprocessing plant and R&D facility pools rise corresponding to receipt of fuel shipments, but are controlled at levels between 10^{-3} and 10^{-4} μCi/ml most of the time. Filtration and ion exchange are the principal methods for controlling radioactivity.(22) See Table 7 for filter types. At most Canadian and some U.S. pools, the ion exchange bed also serves as the filter. Vacuuming is a third radiation control method, which removes radioactive particles from the bottom of the pool.

EUROPEAN SPENT FUEL STORAGE EXPERIENCE

A recent German communication(27) indicates that after up to five years of pool storage there is no visible indication that problems are developing on stainless- or Zircaloy-clad fuel. Rods have been removed from spacers without problems. Also, there is no visible leaching of exposed UO_2 at defects in the fuel cladding.

Further work is underway at the Gesellschaft Zur Wiederaufarbeitung von Kernbrennstoffen Mbh to periodically inspect and photograph BWR and PWR fuel bundles, to determine whether discernible changes are occurring in the cladding. An experiment also is underway to periodically monitor corrosion of an aluminum canister. Based on observations to date, the investigators do not expect to find substantial corrosion in ten years.

British spent fuel storage experience was summarized by Mr. B. F. Warner at the 1977 Windscale Inquiry regarding fuel reprocessing in the United Kingdom.

Stainless-clad fuel from the Windscale Advanced Gas Reactor (WAGR) was first stored in the Windscale pool in 1963. Pins examined metallographically after five years in the pool showed no evidence that corrosion was occurring in the pool. Zircaloy-clad fuel from BWR, PWR and SGHWR (Steam Generating Heavy Water Reactor) exposures has been stored in British pools, prior to reprocessing. The SGHWR fuel has been stored up to 8-9 years. Post-irradiation exams after four years of pool storage have not indicated evidence of corrosion attributable to pool storage. One Canadian CANDU Zircaloy-clad fuel rod, irradiated to 6600 MWd/MTU and stored in demineralized water for 11 years, gave no indication of corrosion due to pool storage in a detailed metallurgical examination.

A program to monitor pool-stored fuel was instituted in 1974 at Windscale and is continuing. Approximately a dozen stainless-clad and Zircaloy clad fuel rods have been identified for metallurgical exams after up to ∿10 years of pool storage.

SUMMARY OF FUEL POOL SURVEY

The current inventory of stored fuel appears to be performing satisfactorily, based on visual inspections and radioactivity monitoring. Canadian fuel stored 10 years and returned to a reactor gave no indication of degradation. Very few detailed fuel examinations have been directed to definition of fuel performance in pool storage, but current experience suggests that degradation, if present, is occurring very slowly.

Mechanical damage appears to be a minor factor in fuel bundle degradation. Fuel bundles which developed defects in-reactor (previously described) have been stored, shipped and reprocessed without major problems. Special handling procedures, such as water-tight cans have been developed. However, radiation releases from the defects generally are low and usually permit handling of the fuel on the same basis as intact fuel.

The technology for handling spent fuel has developed over ∿35 years. It has been satisfactory in almost all respects. The exceptions include an occasional fuel handling accident (Table 5), corrosion of some fuel pool components (carbon steel and in some cases, aluminum alloys), and occasional leaks in pool liners.

PRELIMINARY ASSESSMENT OF POTENTIAL DEGRADATION MECHANISMS FOR MATERIALS IN POOL STORAGE

The foregoing discussion has defined the materials in fuel pools and fuel bundles (Tables 10 and 11), and the range of exposure conditions (Tables 6, 7, and 12). The principal consideration now is to further examine the expected performance of stainless steel and Zircaloy cladding as radiation containment boundaries. Also important is the integrity of bundle components which maintain the bundle configuration and allow it to be transported, principally tie plates, spacers, and bails (Figures 9 and 10). Of lesser but substantial importance is the continuing integrity of fuel pool handling and storage equipment.

There is no visual evidence that significant degradation of fuel bundle materials has occurred. However, until recently there was little incentive for detailed confirmation, because the intended pool residence times for the fuel were relatively short, to be followed by reprocessing. Now, with prospects of longer pool exposures, there is increased incentive to confirm the apparently successful performance and to define whether any degradation mechanisms may threaten fuel bundles or pool equipment in extended aqueous exposures in the 20-to-100 y time frame.

In the following section, potential degradation processes which require discussion and possible evaluation are examined.

POTENTIAL DEGRADATION PROCESSES-FUEL BUNDLE MATERIALS

Fuel bundle materials are summarized in Table 11 and the designs appear in Figures 8, 9, and 10. Fuel rod cladding integrity is an important aspect of the reactor and fuel pool exposures because it provides the primary containment for the radioactivity generated in the fuel. The cladding failure mechanisms discussed earlier (p. 21) included some originating from the fuel side and some originating from the coolant side during reactor exposures. Any detailed fuel examinations and mechanism considerations must evaluate the potential for failures from either side of the cladding.

Evaluation of Fuel-Side Cladding Degradation Mechanisms

The primary fuel-side cladding degradation mechanisms are:

- localized hydriding from hydrogeneous impurities in the fuel
- fission product attack

Pellet-clad interactions are important in reactor-induced failures, but do not appear to be important at pool temperatures. The prospect for degradation due to tritium-induced helium also is discussed.

Hydriding Effects in Zircaloy

Hydrogen concentrations in the internal atmospheres of high-burnup fuel rods are likely to be low, because hydrogen tends to absorb in the cladding during extended reactor exposures. Relatively high hydrogen pressures may occur in unfailed, low-burnup fuel discharged prematurely due to failure of one or more rods. However, hydrogen isotope diffusion at pool temperatures (30-50°C) is slow,(28) and progressive hydriding in extended pool exposures is unlikely, though not fully ruled out, since low-temperature, galvanically-induced hydriding can occur (next section). Also, a phenomenon called delayed hydrogen cracking has occurred at relatively low temperatures.

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Zirconium alloys form hydrides which have low ductility, providing a phase through which cracks may readily propagate. The phenomenon of delayed hydrogen cracking has caused failures in Zr-2.5 Nb alloy pressure tubes in the Pickering-3 and -4 reactors.(29) No cracking occurred in Zircaloy-2 tubes in Pickering units 1 and 2. On current understanding, the phenomenon appears to require stress intensity factors of greater or equal to 5 ksi root inch. At 75°C, the threshold stress for cracking is ∿80 ksi for Zr-2.5 Nb. There is an increase in crack propagation rate with increasing temperature. The prospects for delayed hydrogen cracking to occur in Zircaloy cladding under pool storage conditions appear to be minimal, based on a stress intensity factor of 5 ksi root inch, in cases where cracks have initiated on the fuel side of the cladding during the reactor exposure.

Of possible concern is the progressive degradation of localized hydride nodules ("sunbursts") formed during the reactor exposures, either by further hydriding at low temperatures or by mechanical degradation at incipient hydride failures during fuel handling in the pool. The fact that no failures of this type have been identified in thousands of postirradiation fuel handling operations is substantial evidence that it is not a significant problem.

Hydriding Effects in Stainless Steel

Hydrogen solubilities for stainless steel are low and hydrogen permeation is relatively high at reactor operating temperatures. Hydrogen-induced failures have not appeared to cause significant operational problems in reactor exposures. There currently is no evidence that fuel-side hydrogen effects on stainless steel are a threat in pool exposures. Many austenitic stainless steels (e.g., 18-8 Ni,Cr) tend to resist hydrogen cracking.

Fission Product Attack

There is strong evidence from examinations of reactor fuel and from laboratory studies that fission products are a major factor in some fuel failures during reactor exposures. Iodine(30-33) and cesium(31,32) are known to cause cracking of Zircaloy in laboratory studies.

The fission product attack requires transport of volatile fission products to the cladding surface in sufficient quantities to react. High stress levels from pellet-clad interactions also appear to be a major factor in fuel rod failures.

Cubicciotti, et al., studied species on the inside surfaces of two fuel rods irradiated to 13,000 and 28,000 MWd/MTU, respectively.(33) The results suggest that fuel rod temperature during irradiation is probably more important than burnup in determining the species and amounts of fission products which reach the fuel cladding inner surface. Several fission product species were identified on the clad surface of the low-burnup, high temperature rod. Cesium appeared to occur as Cs_2UO_4. The expected volatile species was Cs_2O rather than Cs. Investigations now available suggest that activities of free cesium will be low in fuel with moderate burnups. The occurrence of significant liquid cesium (melting point, 300°K) in high burnup fuel appears improbable.

Some fission product attack on stainless steel cladding has been reported under conditions in Liquid Metal Fast Breeder Reactor (LMFBR) fuel rods.(34) In light-water reactor stainless-clad fuel rods fission product attack has not appeared to cause significant fuel failures.

Fission product iodine is removed from contention as a long-term threat to pool-stored fuel by the short half lives of its principal isotopes(3) and by some evidence that even if present, it would not initiate cracking of Zircaloy at temperatures near those in pool-stored fuel.(31)

Liquid cesium has caused Zircaloy to embrittle near room temperature.(31) However, the availability of significant free cesium at the fuel cladding surface appears doubtful.(33)

Other fission product species with potential to embrittle cladding have been observed on the inner surfaces of irradiated cladding, including tellurium, selenium and cadmium.(33) Very little is known about the aggressiveness of these species. However, the stress states in the cladding are thought to be low (see later section), which would minimize prospects of stress corrosion cracking.

There are no clearly identifiable mechanisms involving cladding degradation by fission product attack which are likely to occur under pool storage conditions. However, future fuel rod examinations should be scoped to include definition of possible fission product interactions with fuel cladding. Based on the work of Cubicciotti, et al.,(33) the worst case would involve high-burnup fuel which operated at high fuel temperatures, particularly near the end of the reactor exposure.

Helium Embrittlement

Tritium from ternary fissions absorbs in Zircaloy cladding and decays with a half-life of 12.3 y to helium-3. Helium is known to embrittle metals subjected to high temperatures when helium concentrations are sufficiently high.(35) Tritium concentrations measured in Zircaloy cladding are only $\sim$0.07 wt ppm(33) ($\sim$2 appm), which is also the maximum helium concentration which could occur. The low temperature and low potential helium concentration essentially rules out helium embrittlement as a credible threat to cladding integrity.

Evaluation of Water-Side Cladding Degradation Mechanisms

Potential degradation mechanisms which interact with exterior cladding surfaces are summarized below:

- oxidation
- stress corrosion cracking
- hydriding
- mechanical effects

The aqueous environments and mechanical operations characteristic of fuel pools were defined in earlier sections.

Mechanical damage appears to be a minor effect in fuel handling operations. A few instances of accidental damage have occurred, but the resulting damage involved only a few rods. Least well defined is the damage which occurs during shipping, though there is no evidence of substantial shipping damage in routine inspections.

The Aqueous Corrosion Environments

The fuel pool corrosion environments which were defined in the survey include the following for nuclear fuel cladding:

Stainless steel:	deionized water(a) boric acid(a)
Zircaloy:	deionized water(a) boric acid(a)

(a) Oxygen-saturated

The temperatures are 1 to 10°C above pool bulk water temperatures, depending on the time from reactor discharge. The maximum pool temperature identified in the survey was 49°C. While pool specifications suggest that temporary excursions to 100°C could occur if heat exchange capacity were lost soon after a fuel discharge in a reactor pool, such excursions are highly unlikely.

The radiation environment at the fuel rod surface varies from ∿10^6 Roentgens/hr (R/hr) at high flux locations on fresh fuel, down to 10 to 100 R/hr on fuel after extended pool residence.

Galvanic couples exist in all fuel bundles and frequently between bundles and canisters. Stray voltages may exist; for example, underwater lighting is used at some pools. Other sources may include stray currents carried by pipe penetrations. However, stray electrical currents are likely to be small in pool waters. In fact, no evidence has appeared for corrosion of pool equipment or fuel bundles from stray voltages.

Oxidation of Fuel Bundle Materials

The conversion of cladding metal to oxide occurs so slowly under conditions similar to pool storage conditions that rates are difficult to measure for Inconel, stainless steel, and Zircaloy. Oxide film thickness determinations were made on Zircaloy-2 process tubes installed in the Hanford K-East reactor.(37) On tubes installed without autoclaved films, oxide film thicknesses were estimated by color matching with anodized films. The reactor-formed oxides also were anodized to establish where they were in the color sequences. The estimated oxide thicknesses are summarized below for two Zircaloy-2 tubes, exposed to treated Columbia River water at ∿90°C for the times shown.

	Estimated Oxide Thickness		Oxidation Rate	
	mg/dm^2	μm	$mg/dm^2/year$	μm/y
Tube 2557, 4 y				
in-flux	0.4	0.03	0.1	0.007
out-of-flux	0.2	0.015	0.05	0.003
Tube 2169, 1.75 y				
in-flux	0.25	0.017	0.14	0.009
out-of-flux	0.13	0.009	0.07	0.005

The rates were similar to those measured in a Hanford laboratory study at 100°C. The data also are in reasonable agreement with extrapolations from the temperature range 232 to 316°C, based on an expression from Kass.(38) The oxidation data, while not precise, show self-consistent values. The reactor radiation (including neutrons) increased the rates by a factor of two in each case. The temperature is higher than pool temperatures and the water chemistry is less favorable than that in most fuel pools. Therefore, fuel pool environments probably would be less aggressive than the K-Reactor tube environment. A further conservatism arises from the fact that spent fuel surfaces have oxide films which will likely reduce oxidation rates on Zircaloy cladding in the pools. Thus, even the out-of-flux rates on the K-reactor tubes are likely to be faster than rates on oxidized Zircaloy spent fuel surfaces. Extrapolation of the above data to 100 years suggests only 5 to 7 mg/dm^2 (0.3 to 0.5 μm) of oxide growth, corresponding to only 0.05 to 0.07% conversion of the Zircaloy cladding to oxide. Even without the conservatisms indicated above, oxidation is a negligible threat to Zircaloy cladding integrity.

Other investigators(39) have pointed out that extrapolation of Zircaloy corrosion rates at 300°C out to 100 years indicates that only about 10 percent of the cladding would be oxidized, again indicating the excellent oxidation resistance of Zircaloy.

The low-temperature aqueous corrosion of aluminum-base, copper-base and iron-base materials has been reviewed.(40) Data by Brush and Pearl(41) indicate that at temperatures below 93°C, stainless steel corrosion rates in flowing (1.8 m/sec) water with 200 $\mu g O_2/\ell$ (200 ppb) are <0.1 mg/dm^2/mo. This rate extrapolates to <120 mg/dm^2 in 100 years, or <1.5 μm of cladding penetration, representing less than one percent of the cladding thickness. Oxygen concentrations are higher in pool waters than in the experiment cited above, but the data of Brush and Pearl indicate that corrosion rates decreased when oxygen increased from 3 ppb to 200 ppb. Whether the higher oxygen concentrations in pool waters (∿5-8 ppm) will provide further benefit or will be deleterious in long-term pool exposures is not clear, but the data suggest that oxidation of the cladding by normal aqueous corrosion is not a likely degradation mechanism for stainless-clad fuel. Inconel generally is slightly more corrosion-resistant than stainless steel.

Jones and Newman(42) conducted tests on oxidized and as-received Advanced Gas Reactor (AGR) fuel cladding materials[a] under simulated water pool storage conditions, including deionized water and boric acid pool chemistries. Pool storage conditions were indicated to be: up to 3 ppm Cl^-, with possible excursions to 10 ppm if heat exchanger leaks developed; temperature, 40°C (104°F); pH 5.2 to 5.8, controlled by CO_2 from the atmosphere.

The corrosion results on the oxidized materials were not prototypic of corrosion on stainless-clad light water reactor (LWR) fuel. The AGR materials were oxidized at 800°C and developed chromium depletion in the metal substrate, apparently accounting for relatively poor corrosion resistance, compared to as-received (unoxidized) AGR materials. Irradiated Windscale fuel elements, oxidized at lower temperatures, do not show significant corrosion in pool storage. Preliminary comparisons between results from irradiated and unirradiated specimens suggested that radiation did not have a significant effect on the corrosion behavior.

Boric Acid Pool Chemistry

Cohen has summarized the use of boric acid for chemical shim in PWR primary system.(43) He gives solubilities for boric acid in water. Over the nominal fuel pool temperature range, the values vary from 11,800 ppm B (30°C) to 20,000 ppm B (50°C), compared to boron concentrations of 1500 to 2500 ppm in pool waters. Over the fuel pool temperature range, there is little change of pH with temperature for boric acid solutions.(43) Studies of boron incorporation in corrosion films are reported, both at high(43) and low (43,44) temperatures. Cohen discussed briefly impurity removal by ion exchange in systems containing boric acid.(43)

Sensitized Types 304 and 316 stainless steel developed stress corrosion cracking in borated solutions (3000 ppm B) at pH's of 4.5 to 7.5 and chlorides in the range of 5 to 200 ppm at temperatures of 80 to 140°C.(13) Decreasing pH and increasing chloride increased susceptibility to stress corrosion

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(a) The AGR alloy is 20/25/Nb steel, with the following nominal composition: 20 Cr, 25 Ni, 0.01 to 0.07 C, 0.75 Mn, <0.7 Nb.

cracking. Sammarone performed experiments with several galvanic couples in boric acid solutions at 50 to 300°C.(45) Approximately 50 dissimilar metal couples were investigated. The only couples which showed evidence of galvanic corrosion were those involving aluminum, 4340 carbon steel, boronated stainless steel, boral, and nickel-plated 80 Ag-15 In-5 Cd.

Corrosion and boron absorption experiments were performed on several metals under irradiation in boric acid solutions (up to 50 ppm B) at ∿50 to 60°C.(44) Pilot plant and laboratory tests also were conducted. The results indicated that carbon steel was not an acceptable material. Some pitting occurred on 6061 aluminum alloy specimens; behavior of 17-4 PH steel was acceptable. Aluminum corrosion was lower in the boric acid solutions than in pure water. No effects of oxygen at concentrations below 1.6 ppm were observed; tests at higher oxygen concentration were not conducted. The tests therefore differ from boric acid fuel pool chemistries, due to lower oxygen and boric acid concentrations.

Effects of Radiation on Corrosion

Effects of reactor radiation(a) on the corrosion of fuel bundle materials have been characterized.(46) At reactor primary system conditions, nuclear radiation has major effects on Zircaloy corrosion in oxygenated coolants, including BWR primary system chemistry. This results in 15 to 25 μm of uniform oxide on high-exposure Zircaloy BWR fuel rods, but with local thicknesses up to ∿150 μm.(47) On PWR Zircaloy-clad rods, oxides on the coolant side are generally 15 to 20 μm at end-of-life,(18) and radiation has only a minor influence on corrosion in low-oxygen water reactor environments. This observation applies to both borated and nonborated reactor coolants.(18,46,48)

Reactor radiation also had minor effects on the corrosion of stainless steel and Inconel-600 in a low-oxygen reactor coolant.(46)

(a) Principally neutron, gamma and beta radiation, with lesser fluxes of alpha, deuterons, etc. The available evidence points to fast neutrons as the principal source of accelerated in-reactor corrosion on the Zircaloys.

In water pool storage, neutron fluxes ($\sim10^6$ n/cm^2 sec through nuclear waste canisters(49)) are almost certainly negligible from the standpoint of corrosion effects. Gamma fluxes on fuel rod surfaces are estimated to be $\sim10^6$ R/hr at reactor shutdown, decaying by 3 to 4 orders of magnitude in four years.

Cowan and Tedman indicate that gamma fluxes change the chemical environments at corroding surfaces, in some cases having a pronounced effect on corrosion processes.(50) In oxygen-saturated fuel pool waters, formation of H_2O_2 will be favored. In some systems, stainless steel potentials are shifted to more passive values. Sensitized materials may shift to a potential regime where the matrix is more passive, but the chromium-depleted grain boundaries remain active, promoting intergranular attack. Cowan and Tedman warn that selection of cleaning solutions must be made carefully to avoid subsequent accelerated corrosion under irradiation. This consideration could be important if processes to remove crud from the fuel become desirable.

There is some evidence that pure gamma fluxes may accelerate Zircaloy aqueous corrosion in the thin film range at elevated temperatures, but the preponderance of evidence suggests that gamma fluxes do not significantly influence Zircaloy corrosion at reactor temperatures.(51) Visual observations suggest that gamma fluxes also do not have an accelerating influence on corrosion under pool storage conditions,(42) but we are not aware of systematic confirmatory data. The situation is similar for stainless steel, viz., lack of evidence that gamma fluxes either promote or inhibit corrosion, but with minimal detailed investigation of interactions between the gamma fluxes and other pool storage parameters such as water chemistry.

Biological Corrosion

Bacteria and other biological species are known to cause corrosion of numerous materials including iron-base alloys. There does not appear to be evidence that zirconium alloys are susceptible to biological corrosion. Some algae growth occurs in fuel pools, particularly on concrete walls when water chemistry is not carefully controlled. Algae growth has been a minor problem in pools with stainless steel liners which maintain high water quality, as indicated earlier.

High radiation fields and lack of nutrients appear to mitigate biological growth on fuel bundle surfaces. However, some biological species have shown substantial tolerance to radiation,(52,53) and in fact tend to develop resistance with mild radiation exposures.

Beyond some troublesome algae growths, there does not appear to be documented evidence that biological effects have been a problem in fuel pool storage operations.

Effects of Crud Layers

The range of crud layer thicknesses varies from almost nil up to ∿150 μm. Compositions also vary, depending on the materials in the primary circuit. The principal crud species are oxides of iron, nickel and copper and spinels (mixed oxides).

The crud layers are themselves generally inert to the fuel bundle materials. They could influence corrosion by formation of differential aeration cells.(56) Such cells operate by local depletion of oxygen in or under deposits, leading to electrochemical potentials between areas of high and low oxygen concentration. Relative oxide stabilities suggest that Zircaloy may be less susceptible to this type of attack than the stainless steels.

A second possible effect of crud layers is to concentrate aggressive species such as chlorides. There is some evidence that alumina corrosion products can concentrate chlorides.(57) Scrutiny of existing crud analyses and selected additional analyses may determine whether the corrosion environments in crud layers are as inert as they currently are regarded to be.

Residual Stresses in Irradiated Fuel Cladding

The residual stress levels in fuel cladding are expected to be low, based on the following considerations:

- the reactor exposure tends to relax high stresses from fabrication(54)
- upon cooling from reactor operating temperatures, the fuel tends to shrink away from the cladding, minimizing interfacial contact pressures due to pellet-clad interactions at pool temperatures

- residual gas pressures are relatively low, but do account for some residual stresses, particularly in pressurized fuel rods (see Appendix A).

Some cases of pellet-clad bonding have been observed on high-burnup fuel.(55) Clad creep-down onto the pellet column also frequently occurs, particularly involving PWR rods. In some cases, fuel densification has offset some of the loss of pellet-cladding gap due to cladding creep-down. BWR fuel has been generally less susceptible to clad creep-down, due to lower fast fluxes and thicker clad walls.

A preliminary analysis has been made to assess fuel clad stresses due to residual gas pressures in the fuel rods at the end of the reactor exposure (Appendix A). The analysis was conducted using the GAPCON I computer code. It accounts for residual pressures from fill gas (helium) and from fission gas.

For BWR fuel rods the calculated end-of-life gap pressure is typically near 30 psi. This results in a maximum clad stress of less than 500 psi. Occasionally, the gap pressure reaches 75 psi, and rarely it reaches 200 to 300 psi, resulting in corresponding hoop stresses of 1000, 3000 and 4000 psi, respectively.

For PWR fuel, the end-of-life gas pressures in pre-pressurized fuel are typically 250 to 550 psi, causing hoop stresses of 4000 to 8000 psi. Occasionally, PWR gap pressures reach 800 psi, with a corresponding hoop stress of 11,000 psi. The rare maximum PWR gas pressures are ∿1200 psi, resulting in a hoop stress of approximately 17,000 psi.

The above stresses compare to room temperature yield strengths of 70,000 to 100,000 psi for irradiated Zircaloy.(6) In the worst case, clad stresses due to residual gas pressures correspond to slightly over 20 percent of the room temperature yield strength. More typically, they will be five percent or below for BWR rods and ten percent or below for PWR rods.

Stress Corrosion Cracking

The arguments that stresses in fuel cladding are expected to be low were presented in a previous section. There is nothing in current fuel storage experience to suggest that stress corrosion cracking is operative. Any residual concerns, though slight, revolve around the fact that some stress-assisted corrosion phenomena can develop at relatively low stress levels. Also, local stresses, e.g., possibly generated during fuel shipments, have not been characterized.

Stress Corrosion of Austenitic Stainless Steel

Stainless steels, particularly in the sensitized condition, are susceptible to stress corrosion cracking in a variety of environments. Sensitization occurs in a temperature regime which causes chromium carbides to precipitate in grain boundaries (e.g., a few minutes at 600°C is sufficient to cause sensitization). Weld zones frequently have some degree of sensitization. On the other hand, solution-annealed austenitic stainless steels are not susceptible to stress cracking in sea water (∿3 wt% Cl^-), even at high stress levels, in the temperature regime below 65°C (150°F).(58)

Some investigators have observed cracking in austenitic stainless steels in chloride solutions at less than 100°C(50,59) though there also is contrary evidence.(60) Theus and Cels reported that stainless steels have cracked in fluoride solutions at concentrations near one wt ppm.(61) They reported that sensitized austenitic steels have cracked in dilute aqueous fluoride solutions at 65°C.

Low-carbon stainless steels (L grades) and stabilized alloys decrease the prospects that cracking of fuel bundle components will occur during pool residence. Stainless steel end fittings (nozzles and tie plates) generally are fabricated by casting, which also mitigates prospects that stress cracking will occur. Our survey has not revealed any cases of bail, tie plate or bolt failures on commercial pool-stored fuel.

Stress Cracking of Zirconium Alloys

Zirconium alloys are generally regarded as being relatively immune to stress corrosion cracking in aqueous media. Speidel has made an extensive review of zirconium alloy cracking tendencies in numerous environments.(62) He indicates that zirconium alloys are immune to stress corrosion cracking in air, steam, water and in many aqueous solutions. Cracking has been observed under certain conditions in aqueous halide solutions, organic liquids, liquid metals, halogen vapors and hot or fused salts. This must be kept in mind in any cases where it may be necessary to subject a spent fuel bundle to a cleaning process. Cox found that unnotched Zircaloy will crack in neutral aqueous solutions containing 0.005 to 5.0 wt percent chloride ion at ∿25°C with stresses near the yield point,(63) if the specimens are polarized anodically beyond the film breakdown potential. Relations of this environment to expected environments in the fuel pools are remote. Similarities between the behavior of titanium and zirconium alloys suggest that the corrosion behavior of titanium may be potentially instructive for identification of some cautionary regimes applicable to zirconium alloys,(64) if due note is taken of differences in cracking tendencies of α, $\alpha+\beta$ and β titanium alloys.

Galvanic Corrosion

The galvanic couples which exist within fuel bundles and between fuel bundles and other fuel pool components are not expected to present problems, with the possible exception of contacts with aluminum alloys.

The generally passive nature of the stainless steels permits coupling to other passive materials such as Inconel and Zircaloy without expectation of significant corrosion due to the couple. Stainless steel performs best in aerated or oxidizing conditions, which help to maintain the passive state.(57) Boric acid solutions have resulted in galvanic corrosion on some dissimilar metal couples, including those which involve aluminum alloys or carbon steel as one member.(45)

Stainless steel can be coupled to aluminum alloys in environments where aluminum remains passive. This appears to be the case in most, though not all pool environments. Aluminum racks were replaced with stainless steel at one pool, due to corrosion near an aluminum weld. Aluminum alloy canisters corroded in contact with stainless steel in a pool with boric acid pool chemistry. However, at a BWR pool, no galvanic corrosion was observed at contacts between aluminum canisters and the stainless steel pool liner in an inspection after 7 years of pool operation. Insulators and aluminum anodizing have been used to reduce the effect of the galvanic couple. An example of aluminum degradation in a pool with high chloride content was cited earlier. However, in large measure aluminum appears to perform satisfactorily in pools, including components galvanically coupled to other metals.

The influence of aluminum on hydriding of zirconium alloys is a potentially important consideration which is discussed in a later section.

Crevice Corrosion

Crevice effects are also a pertinent consideration, because of numerous crevices in fuel bundles and fuel pool components. Concentration cells promote pitting due to electrochemical action. However, Inconel, stainless steel and Zircaloy appear to resist crevice attack under pool storage conditions. Some uncertainties exist for materials exposed to low pH environments, particularly boric acid in the pool storage case. Aluminum is the most susceptible of the fuel storage materials to crevice corrosion. Results cited earlier indicated that in fact some crevice corrosion has occurred on aluminum canisters in a fuel pool having boric acid chemistry. However, no crevice corrosion was observed in aluminum/stainless steel crevices after 7 years in a BWR pool (previous section).

Galvanically-Induced Hydriding of Zirconium Alloys

There is ample evidence that titanium and zirconium materials will undergo severe hydriding at temperatures less than 100°C when cathodically polarized, either by galvanic couples or by impressed potentials. Severe hydriding occurred on Zircaloy tubes in the Hanford K-reactor at locations where aluminum alloy spacers were in contact with the tubes. Temperatures were $\sim$90°C and the coolant was treated Columbia River water.

Mechanistic studies(66-68) indicated the following characteristics of the hydriding phenomenon:

- Hydride layers up to ∿125 μm thick could develop in a few weeks or months on Zircaloy coupled directly to aluminum alloys or subjected to cathodic dc potentials of 3.0 volts and above;
- aluminum corrosion was essential to anodically polarize the aluminum and cathodically polarize the Zircaloy, causing the Zircaloy to attract and discharge hydrogen
- alternating currents also induced hydriding
- the electrolyte is important to the hydriding; in high purity water, hydriding did not occur over periods of a few months
- hydriding was highly dependent on the metals in the galvanic couple: aluminum alloys 6061 and 8001 both caused Zircaloy to hydride, but the extent was more severe with alloy 6061; anodizing the aluminum largely suppressed Zircaloy hydriding by forming an electrically-insulating oxide film; nickel-plated aluminum coupled to Zircaloy suppressed hydriding and stainless steel couples did not hydride the Zircaloy.
- the surface condition of the Zircaloy was an important factor; grit-blasted Zircaloy was highly susceptible to hydriding; as-etched (pickled) Zircaloy had a much lower hydriding tendency; oxide films formed by autoclaving the Zircaloy prevented formation of hydride layers, though some local hydride needles were observed, presumably forming at imperfections in the autoclaved films;
- grit-blasted titanium hydrided at 55°C in a laboratory test in reactor processes water with impressed potentials;
- recent tests on Zircaloy coupled to aluminum in high purity water did not show evidence of significant hydriding at 20°C and 70°C after one month.(69)

Interpreting the above observations in relation to fuel pool conditions, the following aspects appear to favor low rates of galvanically-induced hydriding:

- relatively high purity waters (low conductivities)
- general absence of direct Zircaloy-aluminum alloy contacts; the contacts usually occur via stainless steel end fittings, though bowed fuel rods may contact canister walls
- oxide films on the irradiated Zircaloy fuel rods
- normal pool temperatures below the range where rapid hydriding has been observed.

Thus, on present knowledge, there are not strong reasons for concern regarding Zircaloy hydriding in aluminum alloy storage canisters. There are some lingering questions, however, regarding the possibility of slow, but progressive hydriding over extended times, where the couple exists.

The results referenced above suggest that Zircaloy-stainless steel couples are not likely to cause accelerated hydriding in pool storage. Zircaloy/Inconel couples have induced rapid hydriding of Zircaloy in-reactor and also in autoclave tests at 280°C.(46) The low fuel pool temperatures seem unlikely to cause rapid hydriding at the Inconel/Zircaloy contacts. In fact, Inconel spacer contacts with the fuel rods have survived reactor exposures without evidence of deterioration.

Pitting Corrosion

Pitting corrosion occurs on stainless steels in sea water, particularly if the surface is not kept free of deposits.(49) Easily-reduced ions such as Fe^{+3}, Cu^{++} or Hg^{++} increase the pitting tendency of stainless steels.(70) Aluminum alloys also are susceptible to pitting corrosion and are adversely affected by heavy metals. While analyses for heavy metal ions are not run routinely at most pools, the available analyses indicate values of less than 0.1 ppm, appearing to be below values which would promote attack on stainless steels and aluminum. Some additional analyses may be needed to confirm that low heavy metal concentrations are in fact routine in fuel pool waters.

Corrosion Behavior at Fuel Defects

The survey indicated that metallic uranium fuel is susceptible to failure at reactor-induced defects or post-irradiation mechanical damage. However, the performance of uranium oxide fuel with cladding defects appears to be satisfactory. If any degradation of cladding is underway at defects, it is occurring slowly in the case of oxide fuel.

A potentially important issue is the behavior of fuel with defects if it is transferred to a storage container, for example, for eventual dry storage. The fuel temperatures inside the container may be substantially higher than the pool storage temperatures. Water may have penetrated into the fuel through some fuel defects. At pool temperatures reactions between the water and the fuel appear to occur slowly.(12,27) However, recent Canadian studies suggest that at elevated temperatures (∿300°C) substantial oxidation of the UO_2 can occur, resulting in swelling of the fuel, if an oxidant is available.(39) While not relevant to pool storage, this observation suggests that further evaluation of the numbers and types of defects, the amount of water-logging and its consequences (if any) as a function of temperature need further definition as a basis for defining the performance of defective fuel in dry storage, if that option becomes operational.

CORROSION OF FUEL POOL EQUIPMENT

Materials in fuel pools are summarized in Table 10. With few exceptions, the materials have functioned well. The exceptions identified in the survey include the following:

- replacement of aluminum canisters due in part to corrosion at a weld
- replacement of a carbon steel heat exchanger due to release of rust to the pool when the heat exchanger was put into service after being on standby
- rusting of a carbon steel pipe after one year in a pool
- failure of an aluminum hanger in a pool having high chloride and nitrate concentrations

- mildly accelerated corrosion of aluminum canisters during periods when pool chemistry was not optimum
- pitting and crevice corrosion on aluminum canisters galvanically-coupled to stainless steel in boric acid pool chemistry
- rusting of fuel racks constructed from galvanized pipe

As with the fuel bundles, the assessment of fuel pool equipment performance is based principally on visual inspections. Few examinations to specifically define the status of materials corrosion have been conducted. This is due to the generally low expected corrosion, particularly in pools with well-controlled chemistries, and to the general absence of equipment problems. A corrosion study conducted at the GE-Morris pool indicated corrosion rates on stainless steel in the pool water which were reported as "infinitesmally small".(23)

Aluminum appears to be the material most susceptible to corrosion. However, aluminum forms a protective oxide in many media. Growth and deterioration of that film is the controlling feature of aluminum corrosion. At 50°C in oxygen-saturated, deionized water, the corrosion rate of the 1100 series aluminum alloy is 0.03 mg/dm^2 day.(71) Extrapolated to 100 years, the total metal penetration is ∿1000 mg/dm^2 (∿37 μm). This oxidation rate is much higher than those cited earlier for Zircaloy (5-7 mg/dm^2 or 0.3 to 0.5 μm in 100 y) and stainless steel (<120 mg/dm^2 or <0.05 μm in 100 y) at pool storage temperatures. However, it does not constitute a threat to the mechanical integrity of aluminum components such as canisters. On the other hand, there are some cautions. First, aluminum is more susceptible than other fuel pool materials to corrosion promoted by metal ions such as copper and mercury. Concentrations of these species are low in pools where we are aware of analytical results. However, some pools have copper alloy heat exchangers and other copper materials. Also, aluminum is often more susceptible to crevice corrosion than other pool metals. In oxygenated solutions, crevice corrosion tendencies are enhanced by differential aeration cells.(56) Aluminum alloys have minimum corrosion rates near pH 7 at fuel pool temperatures.(40) The rates rise sharply as pH's increase beyond 8.0-9.0. At pH's of 4 to 5, the corrosion rates generally are low, but again rise sharply at pH 3.0 and below.

Pitting and crevice corrosion of aluminum has occurred in oxygenated boric acid solutions in experiments conducted by B. Griggs at Hanford. Similar behavior has occurred on aluminum alloy canisters exposed in boric acid pool waters in contact with the stainless steel pool liner. Pitting of aluminum has occurred in high chloride waters, but chloride attack will not be a problem at <1 ppm Cl^- normally encountered in spent fuel pools. Anodizing and elimination of galvanic couples will, where they are problematic, diminish aluminum corrosion.

The principal ionic aluminum corrosion product in basic water is AlO_2^-, which is not easily removed by ion exchange.(43) Aluminum corrosion products tend to build up in the water, finally nucleating precipitates, which may contribute to turbidity problems where substantial corrosion is occurring. Concern also has been expressed regarding clarity problems due to aluminum borate formed by corrosion of aluminum canisters in boric acid pool waters. However, we are not aware of any actual cases where aluminum corrosion products have generated serious pool clarity problems.

Another caution in the application of aluminum involves its potential to cause hydriding of zirconium alloys in certain environments and configurations, as discussed earlier. However, there currently is no evidence that hydriding occurs under pool storage conditions.

Despite the cautions indicated above, aluminum has functioned satisfactorily as a canister and rack material in some pools for approximately 15 years. One pool operator who has experience with both aluminum and stainless steel canisters in deionized water believes that either can function satisfactorily with proper water chemistry and metallurgical specifications.

STATUS OF OTHER SPENT FUEL STORAGE OPTIONS

Foregoing discussion has indicated that pool storage currently is the only interim spent fuel storage option. While spent fuel performance in water pools has been satisfactory, other storage options are being investigated. Irradiated fuel bundles from the Douglas Point reactor are currently in a dry storage test module at Atomic Energy of Canada Whiteshell Laboratory.(4) An unfuelled dry storage test module was tested at Hanford,(72) developed under the Retrievable Surface Storage Facility (RSSF) Program.(49)

A program is underway in the U.S. to develop a dry storage concept for spent fuel. An electrically-heated caisson demonstration module is expected to begin operation at Hanford late in 1977. Test modules containing spent fuel also are planned. However, a fully developed dry storage option is still several years away.

ASSESSMENT OF MATERIALS BEHAVIOR IN FUEL POOLS - SUMMARY

The successful performance of fuel bundle materials in pool storage is a major conclusion of this survey. Neither chemical attack (corrosion) nor substantial mechanical damage have developed during pool storage and handling of commercial fuel.

Review of the corrosion literature has not developed major concerns regarding obvious mechanisms which have high probability to degrade fuel bundle materials.

The fact that very few detailed confirmatory examinations have been made on fuel bundle materials after pool storage results from the relatively short pool exposures intended and the belief that no problems were developing. With the prospects of longer pool residence times comes an increasing incentive for specific confirmatory fuel examinations. Based on current perceptions of spent fuel behavior in pool storage, a major initial effort does not appear warranted. Rather, selected, focused, exploratory surveillance appears to be the present need. If evidence of fuel bundle degradation emerges from the initial exams, this may signal the need for broader surveillance. If no evidence is found in the initial exams, only a modest level of surveillance appears necessary to maintain and reinforce the assurances developed in the preliminary exams. Initiation of a surveillance study on spent fuel in a German pool(27) was indicated earlier.

The incentives for fuel exams and surveillance lie: a) in continuing confirmation that wet storage of uncanned spent fuel can be extended as an interim option; b) in the need to define the probable condition of pool-stored fuel when it is removed to either storage in containers or possibly to eventual reprocessing.

The following discussion identifies factors which appear to require further definition, not necessarily with implied priorities:

a) behavior of fuel defects as a function of defect type, cladding type and storage conditions

b) possible effects of pool temperature and water chemistry transients on the subsequent condition of stored fuel, as a function of material and pool chemistry

c) effects of boric acid pool chemistry on materials behavior

d) effects of galvanic couples on the hydriding of zirconium alloys

e) the degree of mechanical damage to fuel bundle materials in spent fuel shipments

f) the incidence and consequences of high residual stresses in fuel bundle materials

g) the possibility of progressive fission product attack on fuel cladding inner surfaces

h) definition of special effects such as crud layer environments and crevice corrosion.

The list is already long and does not include all the possible factors in detail. However, several of the questions can be addressed simultaneously in a given fuel exam. It is important to emphasize that the above factors represent low-key concerns. They have potential significance principally in the event that pool storage were to extend into the 20-to-100-year time frame.

To define whether discernible degradation mechanisms are occurring during pool storage, it is necessary to separate as much as possible phenomena which occurred in-reactor from those which developed during pool storage. While there is no basis to assume that discernible degradation has occurred, an essential approach to detect it appears to require a return to fuel bundles which have had prior examinations. Referring to Figure 11, the fuel bundle history includes pre-reactor, reactor, and fuel pool residences. The results of the reactor exposure have been defined for numerous bundles by detailed nondestructive and destructive examinations, to determine dimensional changes, definition of defect morphologies and defect mechanisms, crud layer properties, corrosion and hydriding phenomena, mechanical properties, gas pressures and compositions, and fuel condition. Methodologies for nondestructive(73) and destructive examinations(30,33,48,74) have been reported.

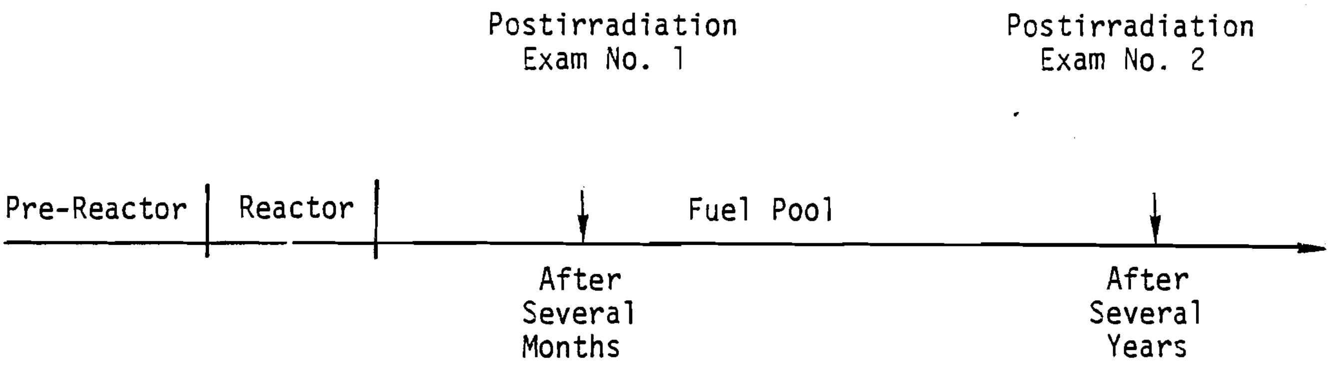

FIGURE 11. Fuel Bundle Exposure Sequence

Several sets of fuel with extended pool residences (up to 18 years) have been identified, having extensive documented examinations to define the phenomena which developed during the reactor exposures. A second set of examinations on the same fuel offers an approach to separate reactor-induced and pool-induced effects, if differences are discernible. Two interesting examples of well-characterized candidates for further exams are: a) Zircaloy-clad Canadian fuel in storage for 14 years in aluminum canisters; b) Zircaloy-clad fuel from the Shippingport reactor, with 13- and 18-year pool storage in stainless steel racks. Other fuel, including stainless-clad bundles, needs to be identified to develop a selection which can provide the maximum information at minimum cost.

As a final observation, the above list of unknowns should not be construed to suggest probable jeopardy for the stored fuel. The satisfactory performance of the fuel has already been stated. The possible degradation mechanisms appear to have low probabilities, but licensing and alternate interim storage issues suggest that further definition of the fuel condition is justified. If fuel bundle material degradation is detectable, it is almost certainly occurring slowly and could be dealt with on a nonemergency basis, either by implementing existing pool storage techniques or their refinements, or eventually by removal of the fuel to storage in containers in a packaged dry or wet storage facility. However, our current understanding indicates that pool storage is an available, viable interim option for control of spent nuclear fuel.

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APPENDIX A

PRELIMINARY ASSESSMENT OF CLADDING STRESSES IN RODS LOCATED IN FUEL STORAGE BASIN

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Introduction

The objective of this work was to find the magnitude of cladding stresses in reactor fuels in spent fuel storage pools. An equation was derived for relating hoop stress to gap pressure and the ratio of cladding radius to thickness. From this equation a stress map of hoop stress versus gap pressure has been developed for typical BWR and PWR fuels. Finally the range of cladding stresses for the fuels is examined.

Hoop Stress Derivation

The largest cladding stresses in nuclear fuels due to gap pressure are hoop stresses on the inner diameter of the clad. The tensile hoop stress due to the gap pressure is:(1)*

$$\sigma_{\substack{\text{max}\\ \text{hoop}_1}} = p\,\frac{b^2 + a^2}{b^2 - a^2} \tag{1}$$

where

$\sigma_{\substack{\text{max}\\ \text{hoop}_1}}$ = inner hoop stress due to gap pressure (tensile)

P = gap pressure

a = inner cladding radius

b = outer cladding radius

The compressive stress at the inner diameter of the cladding due to external pressure is:(1)

$$\sigma_{\text{hoop}_2} = -P_{\text{ex}}\,\frac{2b^2}{b^2 - a^2} \tag{2}$$

---

*See references at the end of Appendix A.

where

σ_{hoop_2} = inner hoop stress due to external pressure (compressive)

P_{ex} = external pressure

a = inner clad radius

b = outer clad radius

Substituting a = r - t/2 and b = r + t/2 in equations 1 and 2 where r is the average clad radius and t is the clad thickness it is found that

$$\sigma_{\substack{max\\hoop_1}} = P\left(\frac{r}{t} + \frac{t}{4r}\right) \tag{3}$$

and

$$\sigma_{hoop_2} = -P_{ex}\left(\frac{r}{t} + \frac{t}{4r} + 1\right) \tag{4}$$

Superimposing the two stresses due to gap pressure and external pressure gives the resultant hoop stress "σ" as shown in equation 5.

$$\sigma = \left(\frac{r}{t} + \frac{1}{4(r/t)}\right)P - \left(P_{ex} + \left(\frac{r}{t} + \frac{1}{4(r/t)}\right) P_{ex}\right) \tag{5}$$

It can be seen from equation 5 that the stress is a function of the gap pressure, external pressure and the ratio of average clad radius to thickness.

Hoop Stress Map

A stress versus gap pressure map that should apply to most BWR and PWR designs is shown in Figure A-1. The fuel is assumed to be submerged in 15 ft* of water in a spent fuel pool. Therefore, the external pressure is atmospheric plus 15 ft of water head. The range for the ratio of clad average radius thickness covers the range for typical fuels as shown in Table A-1 The gap pressure ranges from zero to above 1200 psi, a pressure rarely seen in pressurized water reactor fuels. Therefore the map should cover the range of maximum stresses in typical BWR and PWR fuels.

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*Water heads at the bottom of the fuel bundle can vary up to ∿48 ft in fuel unloading pits and up to ∿30 ft in fuel storage areas.

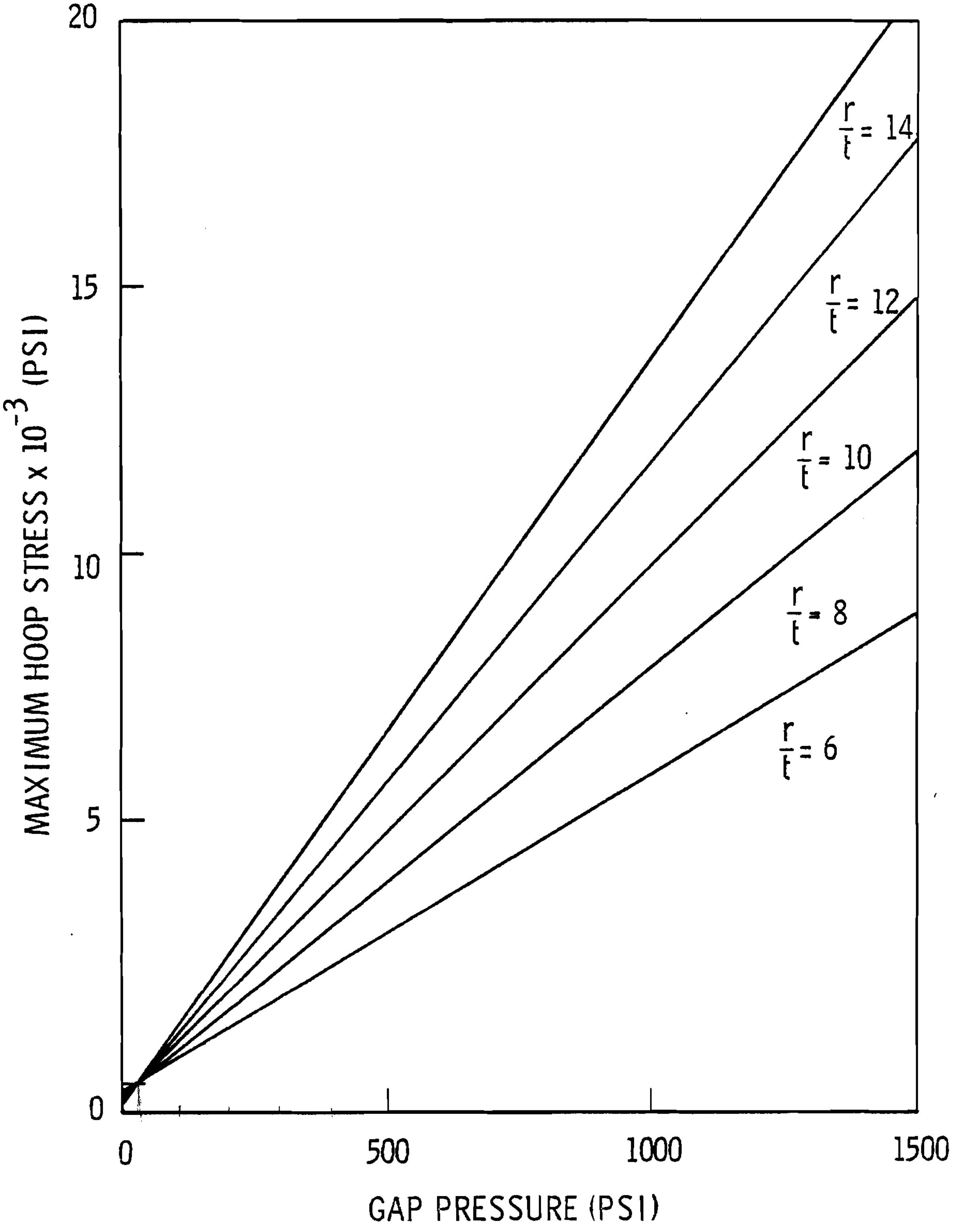

FIGURE A-1. Cladding Hoop Stress Versus Gap Pressure

TABLE A-1. Fuel Dimension Specifications(2)

Contemporary Boiling Water Reactor Fuel

Plant	Clad	Array	I.D. (inch)	O.D. (inch)	Clad Thickness t (inch)	Av. Clad Radius Clad Thickness r/t
Grand Gulf(4)(5)	Zr-2	8x8	0.425	0.493	0.034	6.75
Oyster Creek	Z_r-2	7x7	0.500	0.570	0.035	7.64
Nine Mile Point	Z_r-2	7x7	0.500	0.570	0.035	7.64
Browns Ferry	Z_r-2	7x7	0.498	0.562	0.032	8.28
Easton	Z_r-2	7x7	0.498	0.562	0.032	8.28

Contemporary Pressurized Water Reactor Fuel

Plant	Clad	Array	I.D. (inch)	O.D. (inch)	Clad Thickness t (inch)	Av. Clad Radius Clad Thickness r/t
*Surry 1	Z_r-4	17x17	0.329	0.374	0.0225	7.80
Ginna	Z_r-4	14x14	0.3734	0.422	0.0243	8.18
Kewaunee	Z_r-4	14x14	0.3734	0.422	0.0243	8.18
Turkey Point	Z_r-4	15x15	0.3734	0.422	0.0243	8.18
Indian Point 2	Z_r-4	15x15	0.3734	0.422	0.0243	8.18
Surry	Z_r-4	15x15	0.3734	0.422	0.0243	8.18
San Orofre	S.S.	14x14	0.389	0.422	0.0165	12.3

*Surry 1, 1974 and 2

Analysis

The maximum hoop stresses for typical BWR and PWR fuels can found for various gap pressures by using the clad radius thickness ratios of Table A-1 and the stress map of Figure A-1. The end-of-life gap pressures and corresponding maximum fuel clad stresses are discussed below.

BWR Fuel

The end-of-life gap pressures in current generation BWR fuels are typically ∿30 psi. This results in a maximum clad stress of less than 500 psi. Occasionally the gap pressure reaches 75 psi and it rarely reaches 200 to 300 psi resulting in hoop stresses of less than 1000 psi and 3000 to 4000 psi respectively.

PWR Fuel

The end-of-life (EOL) gap pressures in prepressurized PWR fuels are typically 250 to 550 psi giving hoop stresses from about 4000 to 8000 psi. Occasionally EOL gap pressures reach 800 psi, with a corresponding clad hoop stress of less than 11,000 psi. On rare occasions gap pressures could reach 1200 psi and a corresponding hoop stress of less than 17,000 psi. The non-pressurized PWR rods would yield EOL gas pressures similar to BWR EOL pressures.

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