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EFFECTS OF AN OXIDIZING ATMOSPHERE  
IN A SPENT FUEL PACKAGING FACILITY

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## Abstract

Sufficient oxidation of spent fuel can cause a cladding breach to propagate, resulting in dispersion of fuel particulates and gaseous radionuclides. The literature for spent fuel oxidation in storage and disposal programs was reviewed to evaluate the effect of an oxidizing atmosphere in a preclosure packaging facility on 1) physical condition of the fuel and 2) operations in the facility. Effects such as cladding breach propagation, cladding oxidation, rod dilation, fuel dispersal,  $^{14}\text{C}$  and  $^{85}\text{Kr}$  release, and crud release were evaluated. The impact of these effects, due to oxidation, upon a spent fuel handling facility is generally predicted to be less than the impact of similar effects due to fuel rod breached during handling in an inert-atmosphere facility. Preliminary temperature limits of  $240^{\circ}\text{C}$  and  $227^{\circ}\text{C}$  for a 2-week or 4-week handling period and  $175^{\circ}\text{C}$  for 2-year lag storage would prevent breach propagation and fuel dispersal. Additional data that are needed to support the assumptions in this analysis or complete the database were identified.

# EFFECTS OF AN OXIDIZING ATMOSPHERE IN A SPENT FUEL PACKAGING FACILITY

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## INTRODUCTION

Spent fuel cladding and possibly the fuel pellets may be exposed to an oxidizing atmosphere in a repository packaging facility. Handling operations may be conducted in an air atmosphere, and temporary lag storage may be in an air-cooled vault. Inert atmosphere might be used in canisters for lag storage, but there remains the possibility of a leak or an inadvertent air backfill. In these cases, the outer surface of the Zircaloy cladding will oxidize. Approximately 0.01% of the spent fuel rods will have cladding breaches that occur during irradiation (W083), and additional rods may breach during storage and handling prior to arrival at the packaging facility. Depending on the time at temperature, the  $UO_2$  pellets in the breached rods may oxidize to a higher state.

The  $UO_2$ - $O_2$  phase diagram indicates that the fuel oxidizes to  $U_4O_9$ , then possibly  $U_3O_7$ , and/or  $U_3O_8$  before it reaches its thermodynamically stable state of  $UO_3$ . If moisture is present in the atmosphere, hydration might also occur. As illustrated in Figure 1, oxidation initiates by formation of  $U_4O_9$  on the grain boundaries. The contraction of the  $U_4O_9$  phase causes the grain boundaries to split, providing a pathway for oxygen access to the grains, which then oxidize to  $U_4O_9$  also. As the oxidation progresses, depending on the temperature, low-density phases of either  $U_3O_8$  or amorphous  $UO_3$  form and tear the fuel apart into individual grains. Fission gas entrapped in the fuel matrix is released and the grains themselves start to split. When the low-density

phases form, the expanding fuel places a stress on the cladding with a resultant dimetral deformation. Eventually the initial cladding breach propagates and release of compact powdered fuel particulate may occur. This sequence is slowed as the handling temperatures are lowered.

A long-term program has been conducted at PNL for the Yucca Mountain Project Office (YMPO) to determine the rate of spent fuel oxidation in a potential air atmosphere repository. Atmospheric variables such as temperature and moisture content, along with fuel variables such as burnup and grain size, have been considered. Data generated in this and other programs were used to determine temperature limits that might be imposed in a packaging facility in order to prevent excessive fuel and cladding oxidation. Associated effects resulting from the oxidation such as cladding breach propagation, particulate dispersion, fission gas release, cladding dilation, and crud spallation are assessed to determine possible ramifications for the facility.

#### CLADDING INTEGRITY

A small fraction of the fuel rods will enter the packaging facility with pinhole cladding cracks. As the  $UO_2$  pellets oxidize to  $U_3O_8$  or higher states, a density decrease causes the fuel matrix to expand, thus placing a stress on the cladding. Given enough time, a cladding split will occur at the breach site and propagate. Tests at Argonne National Laboratory (ANL) (EI86) on fuel rod segments indicate, as shown in Figure 2, that the split can take many different forms. In all cases, though, the structural integrity of the cladding is reduced and fuel can be released. Maintaining the fuel temperature below prescribed limits during the various handling and storage steps can prevent the splitting of cladding.

A large number of whole rods or rodlets with cladding breaches were tested in the temperature range of 150 to 360°C to determine the time duration necessary to initiate propagation (see Table 1). Most of the defects were artificial drill holes or slits, but naturally occurring pellet-cladding interaction (PCI) splits were used in some of the CANDU and pressurized water reactor (PWR) fuel studies. Split initiation data for boiling water reactor (BWR) fuel are very limited at all temperatures, and data on PWR fuel are non-existent below 230°C. If the effect of fuel type is not considered, then, above 250°C, fuel oxidation was sufficiently fast that the defects propagated in most of the samples, but below 205°C fuel oxidation was sufficiently slow that most of the defects did not propagate during the tests.

Considering the scatter in the split initiation data (see Fig. 3) and lacking actual formation of splits at the lower temperatures, it was possible only to establish a bounding empirical relationship between temperature and time-to-split initiation. A bounding time-temperature relationship was established that predicted a shorter time-to-split initiation than was actually observed above 250°C and a longer time-to-split initiation below 250°C, where no split initiation occurred during the tests. The activation energy of this relationship is 26.7 kcal/mol; this value agrees quite well with the literature values (BR78, AU58, LA70, MA83) for the diffusion of oxygen in  $UO_2$ , which generally range from 20 to 30 kcal/mol.

Three arbitrary time frames were chosen for this evaluation: 1) 2-week handling duration, 2) 1-month handling duration, and 3) 2 years of packaging facility lag storage. Corresponding temperature limits that would prevent a

cladding breach from propagating into a split are 240, 227, and 170°C, respectively. These temperature limits will vary according to the actual time (including uncertainties) needed to accomplish various steps in the packaging process. Conversely, the facility designer can use Fig. 3 to determine 1) a maximum handling time based on the ability to maintain cooling in the facility and 2) how much the handling must be accelerated if there is a temperature excursion.

If the temperature exceeds the limit for an extended period of time or if the planned time to conduct an activity is exceeded, then the possibility arises that a defect may propagate. The rate of cladding splitting was measured in a limited number of tests. Above 230°C, fuel oxidation starts at the defect site and proceeds down the rod in a front that travels right behind the crack tip at approximately the same velocity (EI85). This is probably due to rapid swelling of the oxidized fuel limiting the oxygen supply to that which can diffuse through the plug of swelled oxidized fuel. There are no data yet to indicate whether the fuel oxidation proceeds in a front at the lower temperatures or if the more rapid oxygen diffusion in the fuel-cladding gap allows the fuel in the rod to oxidize as a unit. Assuming the process is the same at the lower temperatures, both the crack tip velocity and the related oxidation front velocity are plotted in Fig. 4. The propagation velocity,  $V$ , follows an Arrhenius curve, as would be expected from a thermally activated process, described nominally by the equation

$$V(\text{cm/min}) = 4.98 \times 10^6 \exp(-25,043/RT) \quad (1)$$

where R is the gas constant and T is the temperature in K. An upper bound on the propagation velocity would be a factor of three higher. Once again, the activation energy agrees quite well with that for the diffusion of oxygen in  $UO_2$ .

#### CLADDING DEFORMATION

Crack propagation is accompanied by rod dilation. Above 230°C, between 1 to 6% diametral deformation has been measured before cladding splitting occurred, and 1 to 16% after splitting occurred (see Table 2). This deformation is insufficient to cause rod to rod interaction within an intact assembly but may cause handling problems if the assembly is dismantled for consolidation. There is sufficient clearance to pull the deformed rods through the grid spacer springs if there is no splitting. There is a distinct possibility, though, that the grid spacer springs may get caught in a cladding split, either preventing dismantling or breaking a fuel rod. If the split-deformed rods present an operational problem to dismantling, then the temperature limits will have to be lowered.

The Zircaloy cladding is irradiation hardened during the first 10-GWd/MTU burnup. Annealing at some of the higher test temperatures indicated in Table 1 may allow deformation to occur before splitting that would not occur at lower handling temperatures where annealing did not occur. Insufficient data are available to predict the deformation associated with a cladding split that occurs at lower temperatures required for lag storage where the cladding remains in an irradiation-hardened condition.

#### CRUD SPALLATION

Crud and  $ZrO_2$  will spall from the deformed cladding (see Fig. 2) but will not be an operational concern. Assuming that breaches larger than a pinhole

will be removed from the assembly and handled separately, <1% of the remaining rods will have breaches resulting from irradiation or handling. Before split propagation, cladding deformation is localized to a few rod diameters of the defect; i.e., ~0.7% of the rod length. Spallation of 100% of the crud from deformed regions around rod breach sites would be equivalent to <0.01% of the total assembly crud load assuming 1% of the rods were breached either in-reactor or by handling (eventually experience might show the failure rate to be much lower). This is negligible compared to current estimates for crud spallation during normal handling that range from 1 to 15% (SA91). If splitting does occur, then the dispersed radioactivity from the crud will be very small compared to the activity of the dispersible fuel.

#### FUEL DISPERSAL

All the experience with fuel dispersal from split cladding comes from tests above 230°C. Oxidized LWR fuel tends to stay agglomerated within the cladding in aggregates resembling the original fragmented fuel (EI85). Upon handling, though, the oxidized fragments fall out of the cladding and break into a fine powder on impact (EI85, W087, PA87). This particulate consisted essentially of individual grains (see Fig. 5) with a size distribution between 1 and 20  $\mu\text{m}$  (EI85, EI86, W087, PA87, HA86). The size distribution appears insensitive to both oxidation temperature and humidity, at least for advanced gas reactor (AGR) and CANDU fuel (W087, PA87, HA86). As the oxidation state approached  $\text{UO}_3$  in PWR fuel, significant transgranular oxidation occurs (see Fig. 1) (EI86), further reducing the size of the particulate.

Some fuel rods are pressurized during manufacture and others develop an internal rod pressure due to fission gas release while in-reactor. When the fuel rod breaches it relieves the excess pressure and in the process expels



fuel particulate fines from the neighborhood of the breach site. Based on limited data (L080, L081, BU85), roughly 0.003% of the fuel contained in the rod or ~83 mg is released as fines during the breach event. It is expected that some rods will be ruptured in a handling facility even if there is an inert atmosphere. The severity of fuel rod oxidation and conversely the response time needed to limit oxidation can be determined by comparing the amount of oxidized fuel in the vicinity of a cladding split to the fuel released when an intact rod ruptures. As long as the breach has not propagated into a split, any oxidized fuel will be confined within the cladding. Confinement occurs because in-reactor breaches tend to be tight and tortuous. (PA78) and the driving force due to the internal pressurization has been vented. Once the defect propagates, the fuel particulate can potentially escape into the cell atmosphere during subsequent handling.

Depending on the particular fuel rod design, there are between 6 and 11 g of fuel per centimeter for PWR and BWR rods, respectively. Since cracks can propagate up and down the rod with the velocity given in Eq. 1, the weight of fuel,  $W$ , in a cracked section of a PWR rod, that is available for release after a time,  $t$ , is given by

$$W(\text{mg}) = 1.2 \times 10^4 \exp(-25,043/RT) \cdot t \quad (2)$$

where  $t$  is in minutes. This weight of exposed fuel would be ~80% higher if it were a BWR rod and three times as much if the upper bound on the crack propagation velocity was used.

Equation 2 was used to determine the times to expose 83 mg of PWR fuel, an estimated amount released when an intact rod breaches, and ten times this

amount or 830 mg. These are plotted in Fig. 6. Once the time to complete a particular handling step is known, Fig. 3 can be used to determine the maximum temperature to prevent crack extension. If the maximum weight of dispersible fuel allowed in a facility is known, then at these maximum temperatures Fig. 6 can be used to determine the allowable time overrun. For example, if a 2-week handling period was contemplated, fuel temperatures as high as 240°C could be allowed. If one exceeded 2 weeks at this temperature (i.e., because of an equipment failure), then based on Figure 6 it would take only approximately 1 hour for a crack to become large enough to release ~83 mg of fuel (assuming the crack started propagating at exactly the time predicted in Fig. 3). This is equivalent to a rod breach in an inert atmosphere. The time needed to oxidize 83 mg of fuel after 1 month of handling at 227°C or 2 years of storage at 175°C are 2 hours or 1.9 days, respectively. At the maximum allowable handling temperature, the maximum allowable time overrun to prevent 83 mg of fuel dispersal is ~0.3%. This time extension grows as one is willing to accept more fuel dispersal in the hot cell.

Once a crack starts propagating, the only method to prevent fuel dispersal, short of introducing an inert atmosphere, is to decrease the temperature. A drop of ~15°C will double the time necessary to propagate the crack any given distance.

#### CLADDING OXIDATION

There are three possible consequences of cladding oxidation: 1) reduced structural integrity during handling or shortened creep rupture lifetime due to internal pressurization, 2) release of activated material due to flaking of the oxide layer, and 3) release of  $^{14}\text{C}$  or  $^3\text{H}$  entrapped in the cladding.

The extent of Zircaloy cladding oxidation will depend on the availability of oxygen and the oxidation kinetics. Clean Zircaloy will oxidize according to a  $t^{1/3}$  power law until a temperature-dependent transition oxide thickness is reached. In general, the spent-fuel rods attain the transition oxide thickness while in reactor (HI76). After transition, the oxide continues to grow according to a linear rate law. Lower temperature data have been collected, evaluated by Garzarolli, and fit to existing models (GA80). Below 350°C, the model of Dalgaard (DA76) is the most conservative (i.e., predicts the greatest amount of oxidation). The thickness of the Zircaloy that is oxidized,  $\Delta h$ , can be expressed as

$$\Delta h(\text{mil}) = 3.25 \times 10^4 \exp(-22,200/RT) \times t \quad (3)$$

where T is the temperature in K and t is the duration in days.

The expected thickness of the oxidized Zircaloy cladding is plotted in Fig. 7 as a function of temperature for four durations: 2 hours might be typical of an accidental fire, 2 weeks to 2 months as reasonable handling periods, and 2 years as a lag-storage time. Typical cladding thicknesses range from 20 to 35 mil. Even at the highest allowable handling temperatures based on fuel oxidation consequences, cladding oxidation results in a wall thinning of less than 0.05%. If the oxide layer supports no stress, the cladding could be considered to be 0.05% thinner and the stress 0.05% higher. This thinning is within the manufacturing tolerance for the cladding thickness and the increment of additional stress is much lower than caused by the

rod-to-rod variation of the internal gas pressure that causes the stress. Therefore, the minimal oxidation expected should not compromise mechanical stability.

In-reactor performance studies indicate that unlike some crud (corrosion deposits on the cladding) that are known to spall,  $ZrO_2$  (oxidation product of the cladding itself) is very adherent to the underlying Zircaloy cladding. Therefore, release of radionuclides by flaking should not be a problem. Based on recent radial serial etching studies of cladding from a 29.46 GWd/MTU PWR rod (HI90), even if flaking of the oxide occurred, the release from a whole assembly would be ~1.4 mCi. The predominant species would be  $^{60}Co$  with lesser amounts of  $^{54}Mn$ ,  $^{106}Ru$ , and  $^{125}Sb$  (HI90). Alpha release would be many orders of magnitude lower.

#### GASEOUS RELEASE

During irradiation, tritium and  $^{14}C$  form in the cladding and a plethora of fission products, including  $^{85}Kr$ , are formed in the fuel. The tritium and  $^{14}C$  are relatively immobile and remain in the matrix where they are formed. Other products such as gaseous  $^{85}Kr$  can migrate to the grain boundaries and fuel-cladding gap. Under normal operating conditions, fission gas release for PWR fuel is less than 2%, but fission gas release from some of the older BWR fuel has been as high as 50%. This  $^{85}Kr$  is available for release if the rod is breached during preclosure handling. If the preclosure packaging is conducted in an air atmosphere, oxidation may release the volatiles from the fuel or cladding.

The release due to oxidation can be compared to the release due to breaches caused by handling to determine the relative effect of the oxidizing atmosphere. For the purposes of comparison, a 1% breach rate due to handling

will be assumed, although rates as low as 0.2% have been used (JA88). For lack of data and to be conservative, 100% release of volatile radioisotopes from oxidized cladding will be assumed.

#### Gaseous Release from Cladding

A reasonably constant tritium concentration of ~2.7 mCi/g was found near the outer surface of PWR fuel cladding by serial etching (HI90). Approximately ~135 mCi/assembly of  $^3\text{H}$  would be released if 0.05% of the cladding thickness for a whole PWR assembly was oxidized. A release of ~90 mCi/assembly is calculated to be expected from an 8 X 8 BWR assembly that was oxidized (see Table 3). This is a slightly smaller release than the 160 mCi/assembly predicted by Jardine (JA88) when normalized to the same decay time and breach rate.

Measurements of the  $^{14}\text{C}$  concentration in the cladding range from 0.4 to 0.9  $\mu\text{Ci/g}$  Zircaloy for PWR fuel cladding irradiated to ~30 GWd/MTU (GU88, VA91, SM90) and from 0.4 to 1.4  $\mu\text{Ci/g}$  Zircaloy for BWR fuel cladding irradiated to ~27 GWd/MTU (GU88a, VA91). As the cladding oxidizes, some of the  $^{14}\text{C}$  will be released, predominantly as  $^{14}\text{CO}_2$  (SM90). Short-term tests conducted in air (SM90) favor release of  $^{14}\text{C}$  by diffusion from an oxide layer of finite thickness. As much as 47  $\mu\text{Ci}$  of  $^{14}\text{C}$  per assembly can be released if the handling period is long enough so that all of the  $^{14}\text{C}$  is released from the 0.05% of the cladding that is oxidized (see Table 3). If 3500 MT of spent fuel is handled each year (JA88), potentially 0.3 Ci of  $^{14}\text{C}$  could be released in the handling facility. This is less than 5% of the release from a nuclear power plant (PE91), and approximately 6% of the release from rods breached during preclosure handling (JA88).

### Gaseous Release from Oxidized Fuel

If an inert atmosphere was maintained, the temperature of the fuel would have to rise above 400°C before diffusion alone would be sufficient to release substantial quantities of gaseous fission products (BE87). In an air atmosphere, the amount of gas released will depend on the oxidation state of the fuel. As long as only  $U_4O_9$  or  $U_3O_7$  (which do not disturb the lattice structure) form, only the minimal gas trapped at the grain boundaries will be released due to crystal shrinking. After formation of appreciable  $U_3O_8$  or higher oxidation states that rearrange and open the lattice structure, additional gas trapped in the matrix will be released. The cumulative release of  $^{85}Kr$  and  $\Delta(O/M)$  tracked each other (BE87, WI87) in AGR fuel oxidized above 250°C. Although similar experiments have not been done using PWR fuel, it is reasonable to expect this trend to hold. Even after complete oxidation to  $U_3O_8$ , a large fraction of gas will remain in the fuel matrix. As shown in Fig. 8, above 250°C, AGR fuel (W085) releases between 7 and 30% of the  $^{85}Kr$  inventory upon oxidation to  $U_3O_8$ . There are no data to determine if a similar gas release could be expected from LWR fuel. No measurements of gas release have been made at the lower temperatures expected for handling or lag storage.

Before split propagation, the oxidation front extends only a few rod diameters from the breach site (EI86, EI85). At this juncture, less than 1.4% of the fuel in the breached rod has oxidized to  $U_3O_8$ . Assuming that in a worst case, 1% of the rods are breached and any oxidized fuel releases 30% of its  $^{85}Kr$ , then less than  $4 \times 10^{-5}$  of the  $^{85}Kr$  in the rods is released. On the other hand, if only 1% of the gas is in the gap and 1% of the rods breach during handling, then  $1 \times 10^{-4}$  of the  $^{85}Kr$  inventory is released. The release of

$^{85}\text{Kr}$  to the cell by fuel oxidation is less than would be released by normal in-cell handling in either an inert or oxidizing atmosphere.

Once cracks propagate, the  $^{85}\text{Kr}$  gas release will be determined by the amount of fuel oxidized. Again, assuming 30% release of  $^{85}\text{Kr}$  upon oxidation to  $\text{U}_3\text{O}_8$ , under the same breach and release assumptions  $8.2 \times 10^{-4}$  of the gas is released for each centimeter of fuel oxidized. Figure 4 can be used to determine the extent of the breach once the time/temperature history is known. For illustration, it is assumed that during the response period 830 mg of fuel (see section on fuel dispersal), which is equivalent to  $\sim 3.7 \times 10^{-2}\%$  of the fuel in a rod (0.13 cm of oxidized fuel), oxidizes. If 1% of the rods were breached, a maximum of  $1 \times 10^{-6}$  of the  $^{85}\text{Kr}$  inventory would be released. This is only 1% of the  $^{85}\text{Kr}$  that would be released from the fuel-cladding gap of rods that are breached by handling breaches in the preclosure facility. A comparison of the  $^{85}\text{Kr}$  release during normal handling, before split propagation, and after split propagation is given in Table 4.

Similar scenarios can be evaluated for the release of  $^{14}\text{C}$ . These are illustrated in Table 5. When the cladding layer oxidizes on 100% of the rods during normal handling, 20 to 45  $\mu\text{Ci}$  of  $^{14}\text{C}$  per assembly can be expected to be released. If it is assumed that 100% of the  $^{14}\text{C}$  is released from the fuel that is oxidized, then between 19 and 42  $\mu\text{Ci}/\text{assembly}$  is released before cladding split propagation, but only 0.5 to 1.1  $\mu\text{Ci}/\text{assembly}$  is released during the response period after a split starts to propagate. This is considerably less than the  $^{14}\text{C}$  released due to a handling-induced breach (JA88). It should be noted, though, that the  $^{14}\text{C}$  released when a rod breaches is contained in the fuel particulate and is not released as a gas.

The effect of gas release due to oxidation, compared to release due to handling-induced breaches, is very dependent on the amount of fuel that is oxidized (time/temperature dependent) and the actual number of rods breached by handling.

#### DATA NEEDS

During the course of this analysis, assumptions were made and shortcomings in the data were identified. While there is every reason to believe, based on related data and phenomena, that these assumptions are valid, the following needs should be addressed:

1. The tritium profiles on BWR and PWR assemblies are assumed to be the same; however, there are no data on BWR assemblies.
2. Only preliminary data are available to indicate that  $^{14}\text{C}$  comes only from the oxidized layer of the cladding.
3. There are no data on time-to-splitting for LWR fuel below  $250^{\circ}\text{C}$  and only limited data on BWR fuel at any temperature.
4. The rate of cladding splitting has been measured on only a limited number of LWR rods and assumed to be equivalent to the propagation velocity of the oxidation front. While this appears to be true at the higher temperatures, no data are available to support it at the lower temperatures ( $<250^{\circ}\text{C}$ ) where diffusion down the fuel-cladding gap may be sufficiently rapid relative to the oxidation rate to allow the complete rod to participate in the oxidation process.
5. The release of  $^{85}\text{Kr}$  from oxidizing fuel is assumed to be the same for both LWR and AGR fuel; however, no rate of release vs  $\Delta(\text{O/M})$  data is available for LWR fuel.



6. How many rods will enter the preclosure facility breached and how many will be breached during handling.

The extent to which these uncertainties need to be addressed will depend on 1) how close the actual time-temperature conditions will approach the limits defined by the analysis, 2) the uncertainty in the handling conditions, and 3) the ability of a facility to accommodate the release of fuel and fission gases within the facility; e.g., length of lag storage or interruption of cooling air flow.

The handling time in a preclosure facility (<2 yrs) is short compared to the time in either monitored retrievable storage (MRS), dry storage cask, or repository disposal. Data can be obtained on a real-time basis. It is only necessary to assure oneself that the worst-case fuel is being evaluated when setting the temperature limits.

#### CONCLUSIONS

A sufficient database exists to estimate the behavior of spent fuel in a preclosure handling facility that uses an air atmosphere. Temperature limits to preclude detrimental oxidation results were established and the effect of exceeding the limits was evaluated. In particular, it can be concluded that

- Extension of preexisting breaches in the cladding can be avoided if the maximum temperature for 2 weeks of handling is less than 240°C; for 1 month of handling, less than 227°C; and for 2 years of lag storage, 175°C.
- If the time at these temperatures is exceeded, the probability is greatly increased that the cladding breach will propagate and possibly inhibit further rod handling due to excessive dilation or weakened mechanical integrity.

- The fuel oxidation that causes the breach propagation will produce large friable agglomerates that will break into a grain-sized powder as it releases from the split during subsequent handling.
- Additional crud spallation due to oxidation of the cladding is insignificant.
- Oxidation of the cladding is insufficient to weaken its mechanical integrity.
- Krypton-85 release due to oxidation of the fuel is small compared to the release expected from rod breakage during handling.
- If a reasonable response time to overtemperature events is established, the  $^{14}\text{C}$  release due to fuel oxidation will be less than the  $^{14}\text{C}$  release due to cladding oxidation and below the EPA release limits.

Additional data are needed to support some of the realistic assumptions used in arriving at the above conclusions. Because of the short times used in a preclosure handling facility, this can be accomplished in real time.

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## TABLES

1. Oxidation Studies on Breached Fuel Rods.
2. Rod Dilatation Caused by Fuel Oxidation.
3.  $^3\text{H}$  and  $^{14}\text{C}$  Release Due to 0.05% of the Cladding Oxidation.
4. Comparison of  $^{85}\text{Kr}$  Release (normal handling vs oxidation of the fuel).
5. Comparison of Gaseous  $^{14}\text{C}$  Release due to Oxidation (cladding vs fuel).

## FIGURES

1. Steps in the Oxidation of Spent Fuel.
2. Modes of Cladding Fracture.
3. Time Required to Propagate Existing Cladding Defects.
4. Cladding Crack Propagation Velocity.
5. Fuel Oxidation into Grains. After sufficient oxidation the spent fuel fragments will separate into individual grains. Turkey Point spent fuel after 5514 hours in an air atmosphere at 250°C. O/M = 2.42.
6. Time Required to Oxidize Specific Quantities of Spent Fuel.
7. Thickness of Oxidized Zircaloy as a Function of Temperature.
8.  $^{85}\text{Kr}$  Released from Fuel upon Oxidation.



TABLE 1. Oxidation Studies on Breached Fuel Rods

<u>Fuel Type</u>	<u>Burnup GWd/MTU</u>	<u>Temperature Range, °C</u>	<u>Number of Samples</u>	<u>Defect Type</u>	<u>Sample Type</u>	<u>References</u>
PWR	13	295	4	760 $\mu$ m drill	Rodlet	EI86
PWR	27	250-360	10	760 $\mu$ m drill 8-25 $\mu$ m split	Rodlet	EI86
PWR	27	230	2	760 $\mu$ m drill	Rod	OL85, EI85
CANDU	~8	~250,300	7	0.5 mm drill 0.8 mm drill slits, natural	Rod	N083, B077 HA84, HA85
CANDU	~8	150	180	3 mm drill	Rod	WA89, WA84
BWR	36	295	1	760 $\mu$ m drill	Rodlet	EI86a
AGR	15.5	240,220	12	1 mm, 2 mm drill	Rod	W084

TABLE 2. Rod Dilation Caused by Fuel Oxidation

<u>Fuel</u>	<u>Temperature, °C</u>	<u>Time</u>	<u>Dilation, %</u>	<u>Crack Propagation</u>	<u>Reference</u>
CANDU	250		8	Yes	HA84
CANDU	400		5-12	Yes	N083
CANDU	400		<2	No	N083
AGR	215-225 240	27 d	0-5.3 0-7.8	No/Yes No/Yes	W084 W084
BWR	229	5962 h	9.8-15.8	Yes	EI85
PWR	250-360		6.5-7.5 ≤1	Yes <sup>(a)</sup> Yes <sup>(b)</sup>	EI86 EI86

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(a) 760 μm defect.

(b) Natural defect.

TABLE 3. <sup>3</sup>H and <sup>14</sup>C Release Due to 0.05% Cladding Oxidation

<u>Assembly Type</u>	<u>15 x 15 PWR</u>	<u>8 x 8 BWR</u>
Zircaloy weight oxidized per assembly, g	50	33.5
Concentrations ( $\mu\text{Ci/g}$ )		
<sup>3</sup> H	$2.7 \times 10^3$	
<sup>14</sup> C	0.4-0.9	0.4-1.4
Release from one assembly		
<sup>3</sup> H mCi	135	
<sup>14</sup> C $\mu\text{Ci}$	20-45	13-47

TABLE 4. Comparison of  $^{85}\text{Kr}$  Release (normal handling vs oxidation of fuel)

	<u>Normal Handling</u> <sup>(b)</sup>	<u>Oxidation of fuel</u>	
		<u>Pre-Split</u> <sup>(a)</sup>	<u>Post-Split</u> <sup>(a)</sup>
Rods involved, %	1	1	1
Fuel involved in release, %	100	1.4	$3.7 \times 10^{-2}$
Gas released from fuel, %	1	30	30
Inventory released, %	$1 \times 10^{-2}$	$4.2 \times 10^{-3}$	$1 \times 10^{-4}$

(a) Refers to time prior to when a cladding breach starts to propagate as a cladding split.

(b) In either an oxidizing or inert atmosphere.

TABLE 5. Comparison of Gaseous  $^{14}\text{C}$  Release Due to Oxidation  
(cladding vs fuel)

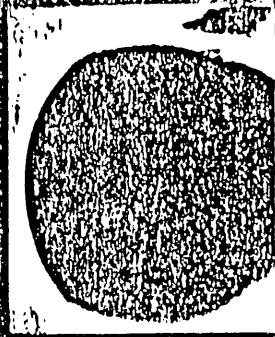
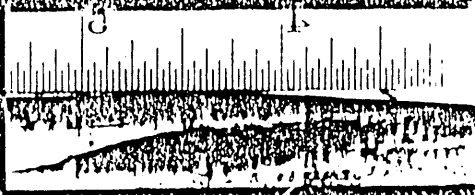
<u>Inventory Ci/MTU<sup>(a)</sup></u>	<u>Cladding</u>	<u>Fuel-Presplit</u>	<u>Fuel-Postsplit</u>
BWR	0.11-.38	0.54	0.54
PWR	0.08-.18	0.60	0.60
Rods participating in release, %	100	1	1
Fuel or cladding per rod participating, %	.05	1.4	$3.7 \cdot 10^{-2}$
Gas released by participating fuel/cladding, %	100	100	100
Release of $^{14}\text{C}$ , $\mu\text{Ci}/\text{assembly}^{(b)}$			
BWR	13-47	19	0.5
PWR	20-45	42	1.1

(a) ~2 PWR assemblies/MTU and ~4 BWR assemblies/MTU.

(b) Average is over all the assemblies handled in the facility.

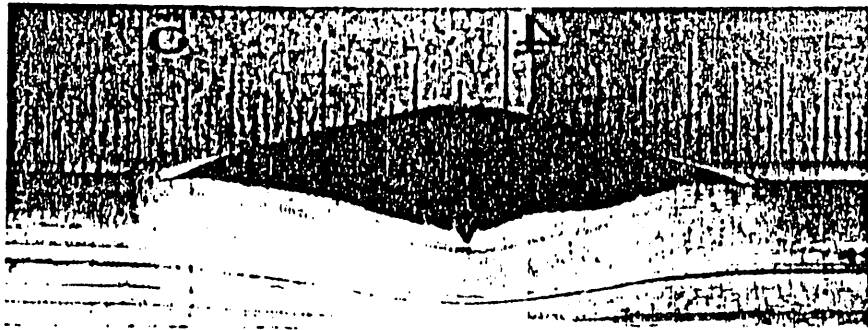
# Oxidation Progression

## Rod Behavior

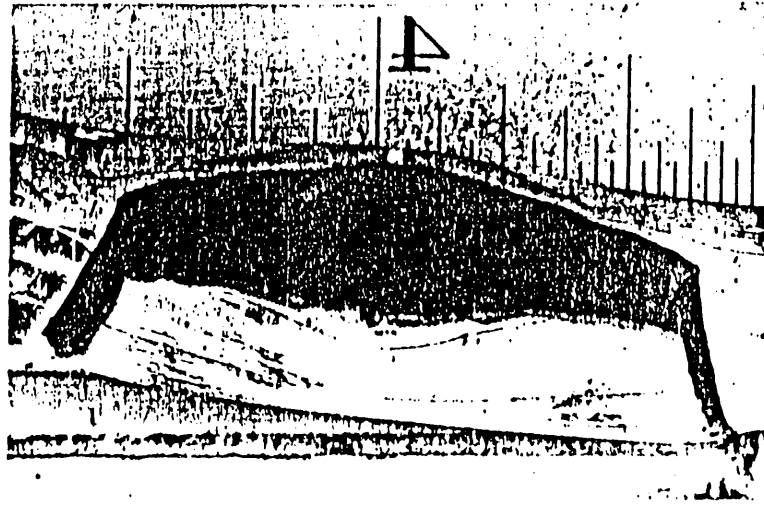


## Fragment Behavior

ORNL-316



a) AXIAL  
360°C  
51.5 HRS



b) FLAP  
325°C  
126.5 HRS



c) SPIRAL  
283°C  
1208 HRS

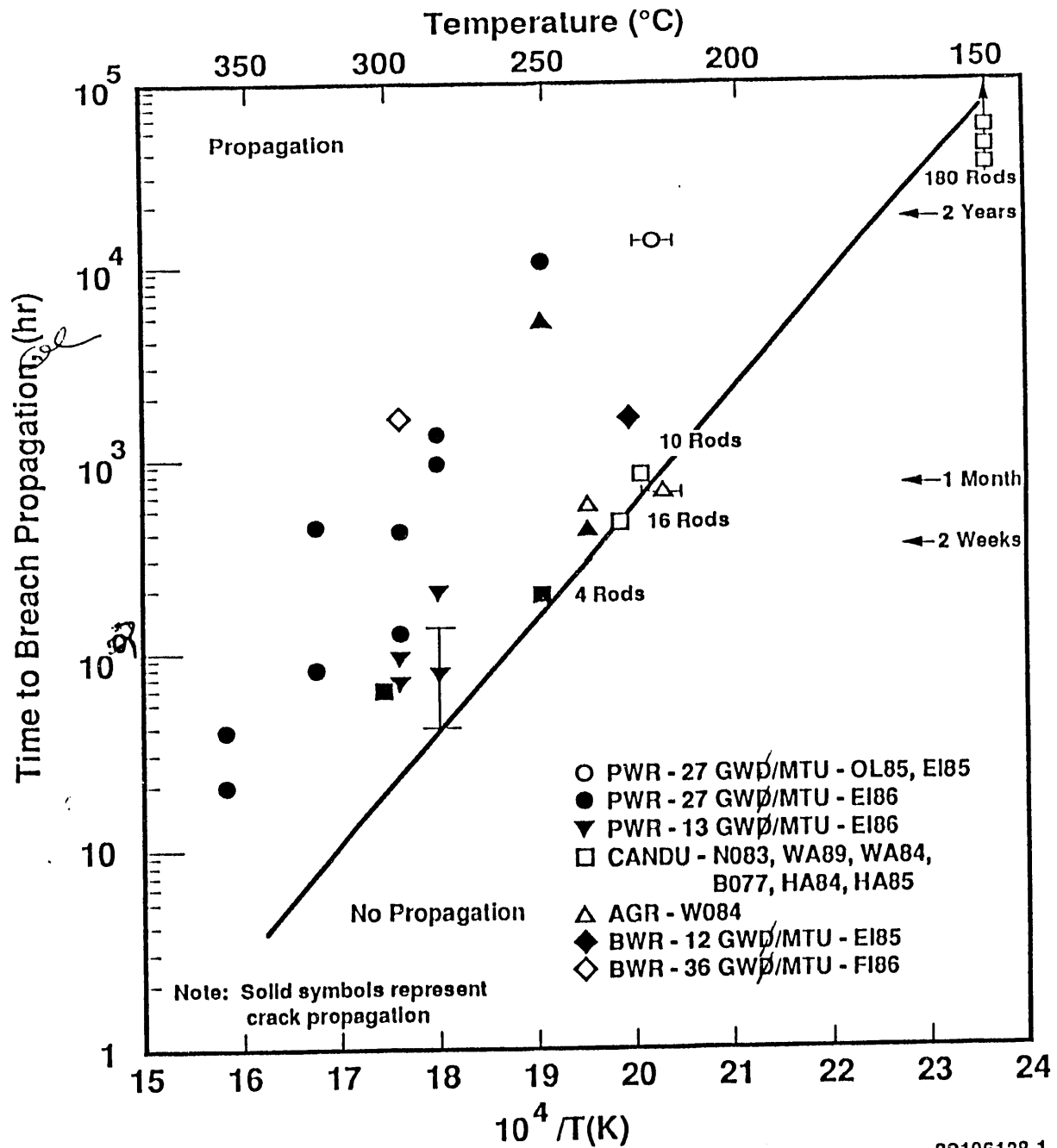
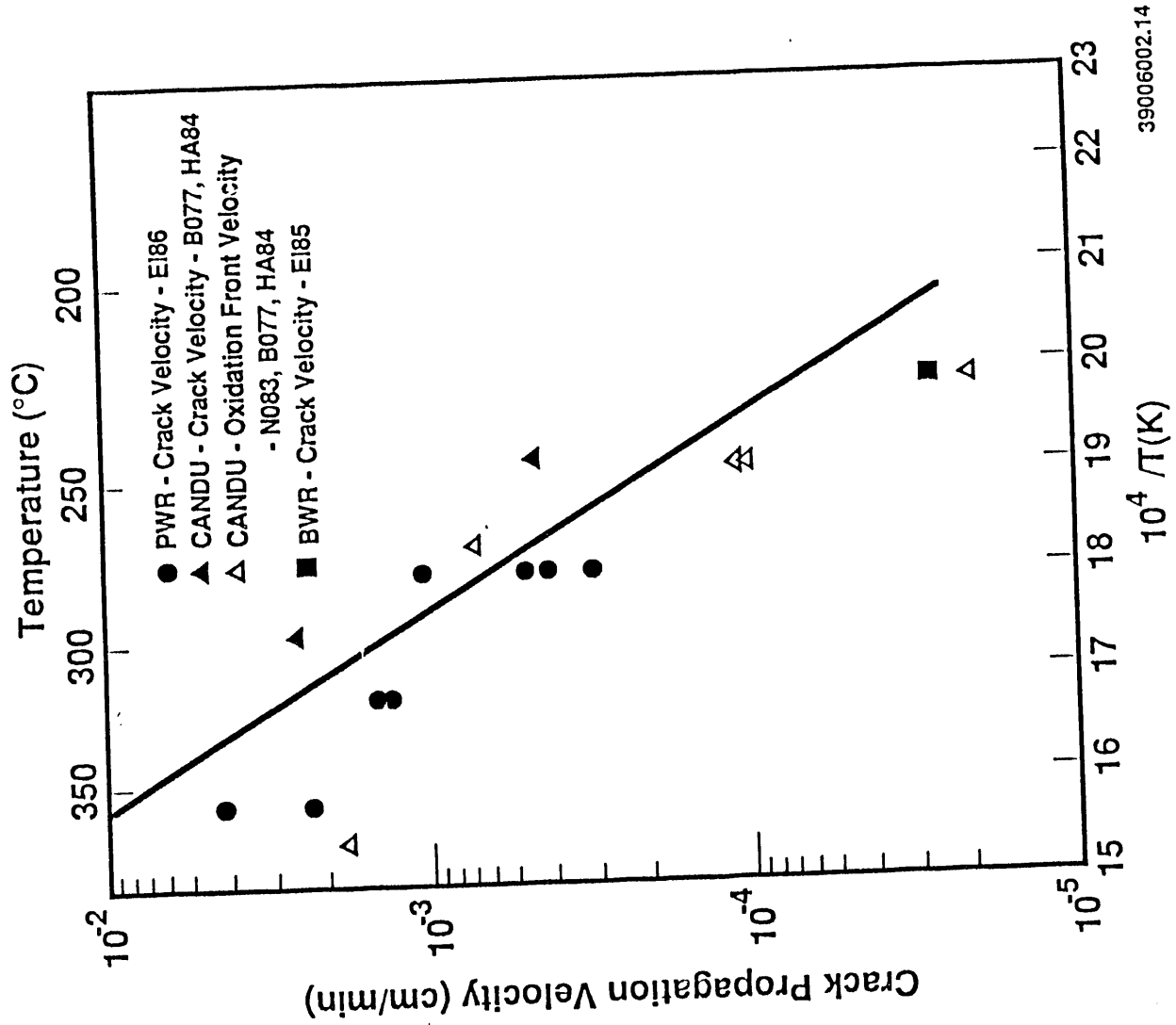


Fig 3





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45E

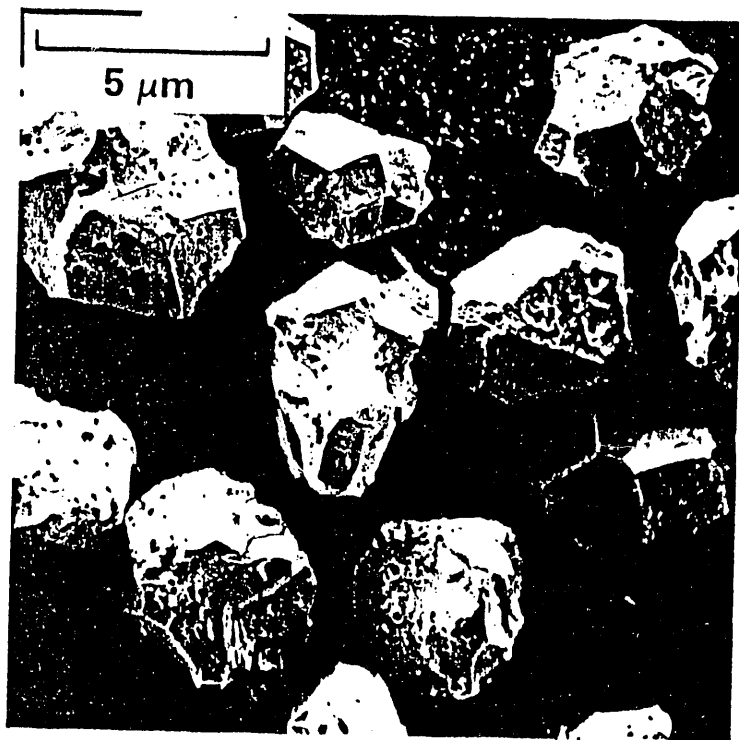
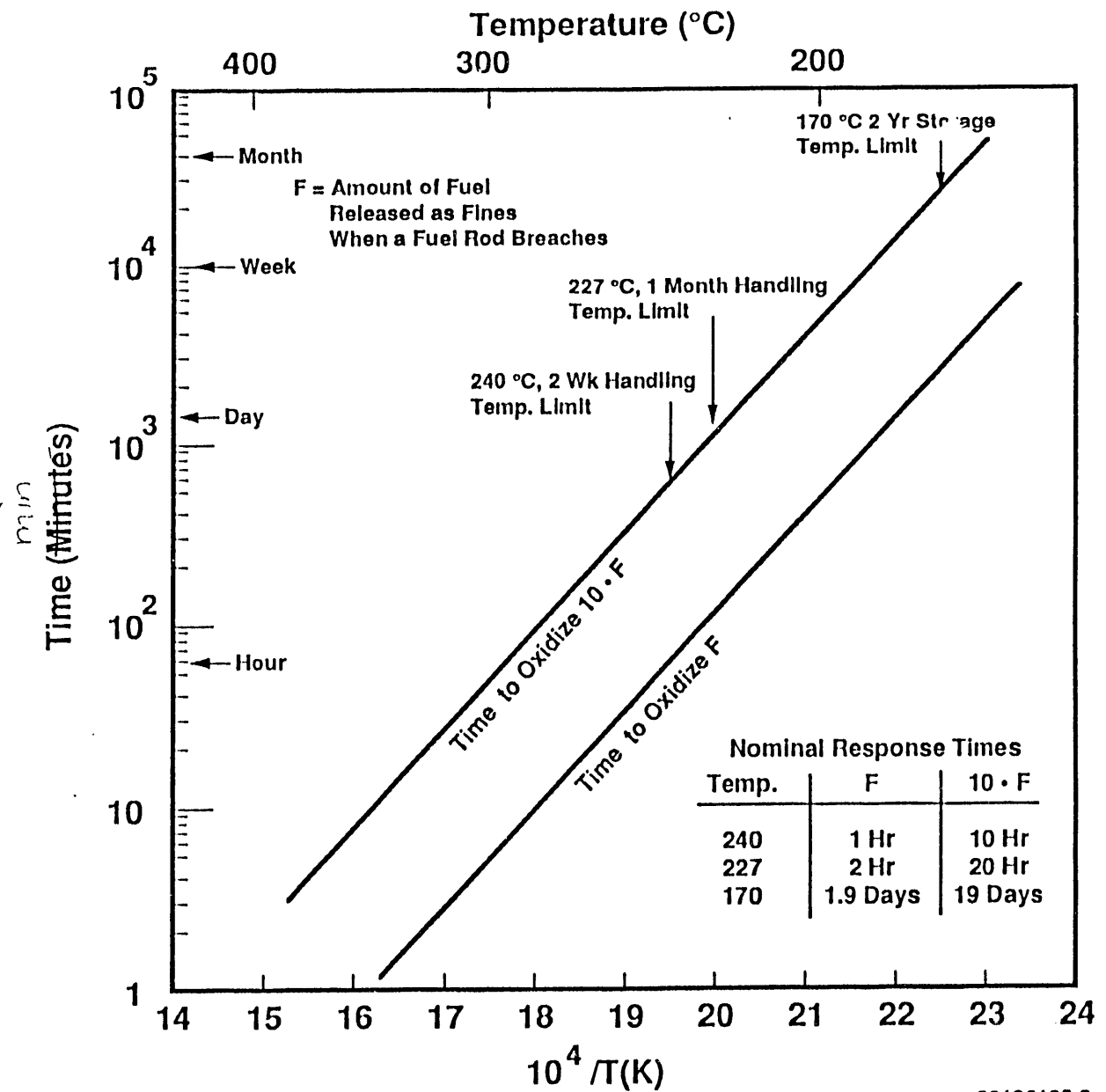
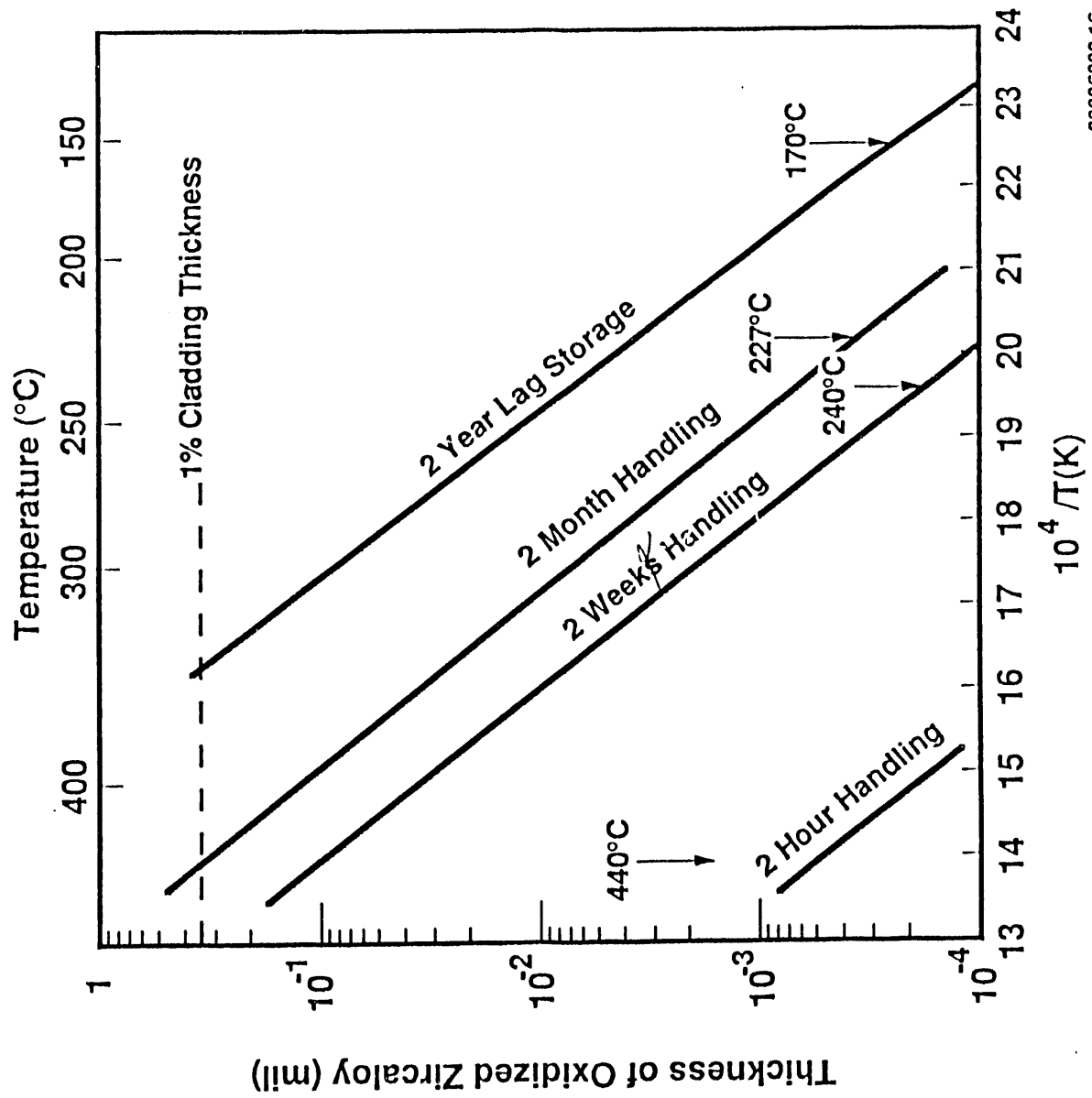


Fig 5



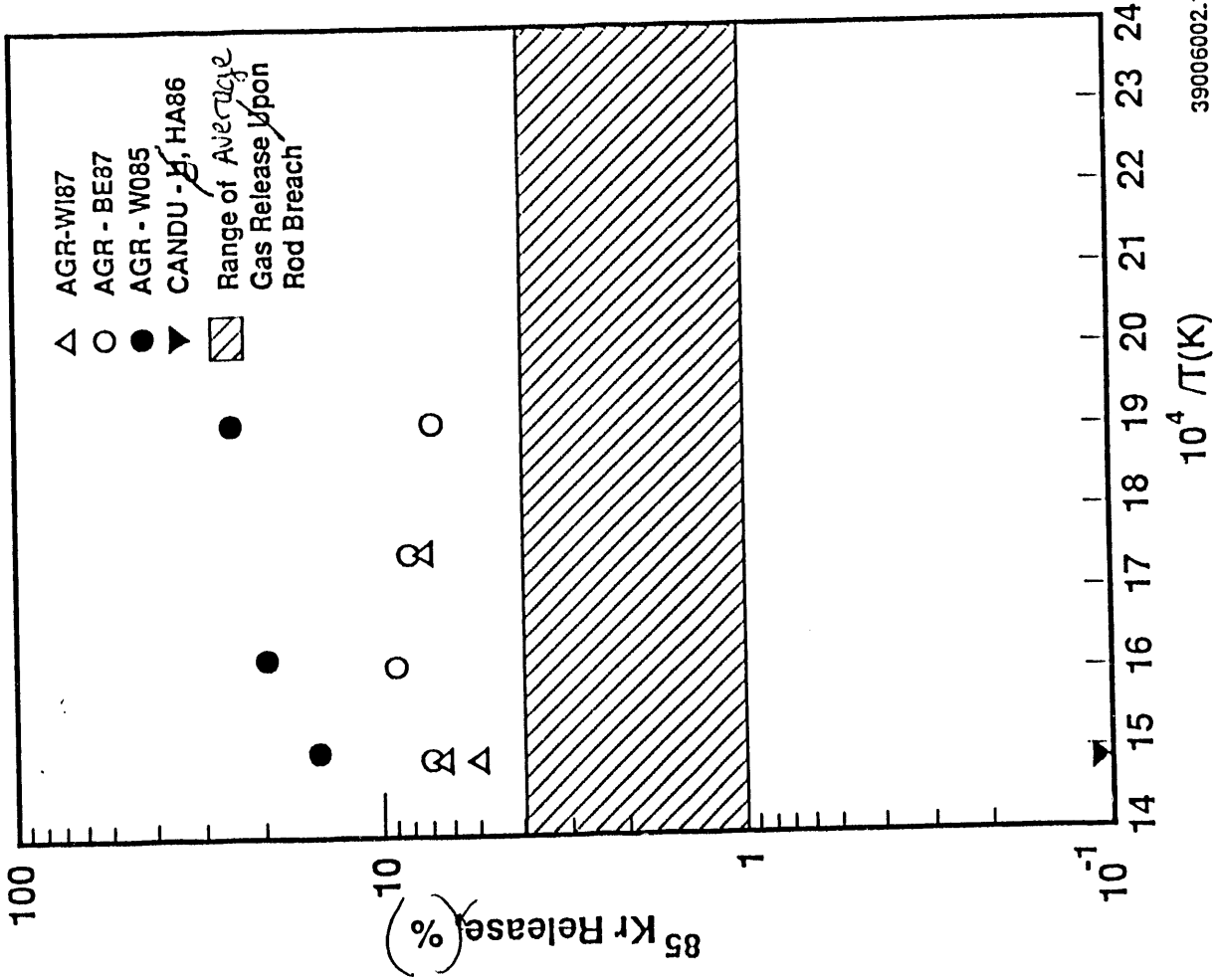
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Fig 6



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Fig 7



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$10^4 T(K)$

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