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**Cesium Chloride
Compatibility Testing
Program Annual Report
Fiscal Year 1985**

G. H. Bryan

March 1986

**Prepared for the U.S. Department of Energy
under Contract DE-AC06-76RLO 1830**

**Pacific Northwest Laboratory
Operated for the U.S. Department of Energy
by Battelle Memorial Institute**



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Richland, Washington 99352

SUMMARY

A program was started in FY 1982 to evaluate the compatibility of Waste Encapsulation and Storage Facility (WESF)-produced cesium chloride (CsCl) with 316L stainless steel (SS) under thermal conditions that may be encountered in a geologic repository. The program is funded through the Defense High-Level Waste Technology Program of the Department of Energy. The principal objective of the program is compatibility testing of six standard WESF capsules at a maximum metal/CsCl interface temperature of $\sim 450^{\circ}\text{C}$. The capsules are placed vertically in insulated containers and allowed to self-heat to the test temperature, where they are maintained for intervals from 0.25 to 5 yr. After the required time has elapsed, each capsule is destructively examined to determine the extent of metal attack by the cesium chloride.

Test capsule No. C-1351 was removed from its insulated container after being held at temperature for 28,268 h (3.2 yr). The aged capsule was shipped to WESF for sectioning. Four ring sections from the inner capsule were returned to Pacific Northwest Laboratory (PNL), where metallographic samples were cut from each ring. Metallographic examination of the samples will be provided by Westinghouse Hanford Company (WHC) in FY 1986.

Control samples of 316L stainless steel, which had been held at 400° , 450° , or 500°C for 28,296 h (3.2 yr), were subjected to metallographic examination. The results show that aging 316L SS at 400°C has very little effect on its microstructure, while aging at 500°C produces extensive grain boundary precipitation. These results confirm the results of shorter duration tests previously reported. The sample aged at 450°C for 28,296 h showed indications of grain boundary precipitation, which was not observed in the shorter duration tests. If continued testing shows that grain boundary precipitation at 450°C continues to increase with time, this could adversely affect the resistance of the 316L SS to attack by the CsCl. This could, in turn, require a decrease in the maximum repository metal/CsCl interface temperature to below the 450°C that is currently under consideration.

Another special study performed in FY 1985 involved the destructive examination of 18 CsCl capsules from the Waste Encapsulation and Storage Facility (WESF) storage pool. The objective was to confirm that corrosion by

the CsCl is not a threat to extended safe storage of the capsules in a water pool. The capsules had been stored in the pool for periods ranging from 0.4 to 8.9 yr. Sectioning of the capsules was carried out at WESF. A ring section from each inner capsule was shipped to PNL for sampling. Metallographic examination of the samples were provided by WHC. The results obtained show that, on an overall basis, corrosion of the 316L SS by the CsCl is low. The corrosion consisted of a general surface attack and pitting, with little indication of grain boundary corrosion.

The maximum attack observed in any of the 18 capsules was about 100 μm (0.004 in.) and in all but two of the capsules the attack was no greater than 50 μm (0.002 in.). There was little correlation between the extent of the corrosion and storage time. Over half of the capsules examined showed no greater attack than was found in previous capsules examined immediately after CsCl filling.

Corrosion was found on the external surface of samples from four of the 18 capsules. This corrosion consisted of isolated pitting. The pitting depth did not exceed about 60 μm .

Evaluation of the corrosion data for the 18 storage capsules examined in FY 1985, and two storage capsules examined in FY 1984, indicates that the CsCl capsules can be stored in water pools for long periods of time without seriously affecting capsule integrity. The external corrosion observed on some inner capsules should not affect safe storage of the capsules

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1.0 INTRODUCTION

At Hanford, fission product cesium, containing 20-40% ^{137}Cs , has been recovered from the high-level waste (HLW) and converted to cesium chloride (CsCl). Recovery of the Cs, as a nitrate, from the HLW and its subsequent purification, takes place in B-Plant. Conversion of the purified product to CsCl , encapsulation of the CsCl , and water storage of the CsCl capsules takes place in the Waste Encapsulation and Storage Facility (WESF). Both facilities are operated for the Department of Energy (DOE) by Rockwell Hanford Operations (Rockwell).

The CsCl is doubly encapsulated in high-integrity 316L stainless steel (SS) capsules. The CsCl is loaded into the inner 316L SS capsules by melt casting. Each inner capsule, which has an ID of 2 in. (50.8 mm) and an inner length of ~19 in. (483 mm), contains up to 3 kg of CsCl . The capsules contain up to ~70,000 Ci of ^{137}Cs , depending on the fission product Cs and the purity of the CsCl .

To evaluate the potential hazards associated with the geologic disposal of the WESF CsCl capsules, reliable estimates of long-term attack of the capsule material by the CsCl at disposal temperatures are required. Available data on the compatibility of WESF-produced CsCl with 316L SS are not adequate for making the required evaluations. The Cesium Chloride Compatibility Testing Program was started at the PNL in FY 1982 to obtain the needed data. The program will take ~6 yr to complete. The work is funded by the long-term High-Level Defense Waste Technology Program of the DOE. This report summarizes the program activities for FY 1985.

2.0 OBJECTIVES

The primary objective of the Cesium Chloride Compatibility Testing Program is to evaluate the compatibility of WESF-produced CsCl with the 316L SS capsule material at the temperatures that could be encountered in a geologic repository. Sufficient short-term (5 yr) compatibility data are to be obtained with WESF-produced CsCl to permit useful estimates of long-term attack on the 316L capsules at potential storage temperatures. A secondary objective for FY 1985 involved the destructive examination of 18 WESF CsCl capsules taken from the storage pool to confirm that corrosion by the chloride is not a threat to continued safe storage of the capsules in a water basin, and to provide an estimate of initial conditions at the beginning of repository disposal.

3.0 TESTING CRITERIA

A number of variables can affect the compatibility of the WESF-produced CsCl with 316L SS in a geologic repository. The more important variables include:

- 316L SS/CsCl interface temperature
- impurities in the WESF-produced CsCl
- changes in the microstructure of the 316L SS due to thermal aging reactions (i.e., precipitation of carbide phases, etc.)
- degree of contact between the 316L SS and the CsCl.

Program scope does not include a detailed testing program to evaluate all of the variables that can affect 316L SS/CsCl compatibility, especially with regard to impurity effects. The limited testing program now underway was designed on the following bases:

- Maximum temperatures in the geologic repository, based on estimates made by Rockwell and PNL personnel, will not exceed 450°C at the 316L SS/CsCl interface.
- The WESF CsCl capsules will be placed in the repository in a vertical orientation.
- The radioactive compatibility tests are to be carried out with standard production WESF CsCl capsules without regard to possible variations in the composition of the CsCl between capsules.

The limitations imposed by the last basis can have a significant effect on the radioactive compatibility data obtained, since theoretical considerations indicate that certain impurities in the CsCl could have a significant effect on the capsule metal attack. Estimates of the impurity content of WESF CsCl, based on ICP results, and an elementary thermodynamic analysis of the 316L/WESF CsCl system have been reported (Fullam 1982). The data obtained in the radioactive tests now underway will provide a measure of the metal attack for a given set of WESF capsules, but will not provide a complete picture of how the attack may vary with changes in the CsCl composition.

The Cesium Chloride Compatibility Testing Program consisted of five tasks.

- Task 1 - radioactive compatibility data lasting up to 32,000 h using standard WESF CsCl capsules aged at elevated temperatures.
- Task 2 - heat transfer studies to define the relationship between the surface temperature of the inner and outer capsules of the WESF CsCl capsule.
- Task 3 - chemical analysis of the CsCl from a batch of WESF-produced CsCl (the CsCl product from WESF is not analyzed, although the Cs feed solution to WESF is analyzed).
- Task 4 - a thermodynamic analysis of the WESF CsCl/316L SS system.
- Task 5 - physical property measurements on CsCl-impurity mixtures.

The major emphasis was and continues to be on the first task. Tasks 2 thru 5 were completed and the results were reported in the annual report for FY 1982 (Fullam 1982).

The program was expanded in FY 1984-1985 to include two additional tasks.

- a study of the pitting observed on the outer surface of some WESF inner capsules
- the destructive examination of 20 WESF CsCl capsules taken from the storage pool.

Work on the first task was completed in FY 1984 while work on the second task involved the examination of 2 capsules in FY 1984 and 18 capsules in FY 1985.

4.0 RADIOACTIVE COMPATIBILITY TESTS

4.1 THERMAL AGING TESTS

The thermal aging tests are designed to provide the short-term corrosion data needed to estimate the long-term attack of 316L stainless steel (SS) by WESF produced CsCl at a maximum metal/CsCl interface temperature of 450°C. The results obtained from the tests should meet this requirement within the limitations described in the previous section.

In the thermal aging tests, six standard WESF CsCl capsules are placed vertically in individual insulated containers and allowed to self heat to a maximum metal/CsCl interface temperature of 450°C. The capsules are maintained at temperature for nominal times of 0.25, 0.5, 1, 2, 3, and 5 yr. The original schedule called for the last capsule to be thermally aged for 32,000 h, but the time has been extended to 44,000 h (5 yr).

When thermal aging of a capsule is completed, it is removed from the insulated container, cooled, and sectioned. Four samples are cut from the inner capsule at specific locations for metallographic examination. Complete details of the testing procedures are provided in previous reports (Fullam 1982, Bryan 1983).

The CsCl capsules used in the thermal aging tests are typical WESF production capsules. The capsules were fabricated and filled with CsCl in accordance with all pertinent Rockwell-WESF and DOE OC and OA requirements.

In addition to the thermal aging tests, two typical WESF inner capsules were sectioned and subjected to metallographic examination immediately after filling with CsCl in accordance with WESF specifications. This provides a measure of the metal attack that occurs when the capsule is loaded with molten CsCl. The two "zero-time" capsules serve as controls for determining the metal attack resulting from the thermal aging. Results obtained from the "zero-time" capsules are presented in earlier reports (Bryan 1984, Bryan 1985) and summarized in Section 4.3 of this report.

Testing and metallographic examination of the first four thermally aged capsules (0.25, 0.5, 1, and 2 yr) has been completed. Sectioning of the capsules and metallographic examination of the metal samples was carried out

at the Oak Ridge National Laboratory (ORNL). Results obtained from the four capsules are presented in Bryan 1984 and Bryan 1985.

Thermal aging of the three-year capsule was completed in FY 1985. The aged capsule was shipped to WESF for sectioning. Metallographic examination of the metal samples from the capsule will be provided by Westinghouse Hanford Company's Post Irradiation Testing Laboratory. Aging of the three-year capsule was originally scheduled to be completed in April 1985. Because WESF could not accept the capsule until July 1985, the capsule was not removed from its insulated container until July 12, 1985. The actual aging time was, therefore, 28,268 h (3.2 yr instead of the scheduled 3 yr). Because of this delay, metallographic examination of the capsule was not completed in FY 1985, as originally scheduled. Results for the 3.2 yr capsule will be presented in the FY 1986 report.

4.2 METAL CONTROL SAMPLES

In many 300 series stainless steel, carbide precipitates form at the grain boundaries when the alloys are held within the sensitizing range (about 400° to 900°C) for extended periods of time. The precipitate formation lowers the corrosion resistance at the grain boundaries. The low carbon content of 316L SS reduces the potential problem but does not eliminate it.

When the 316L SS is held in contact with the WESF CsCl at temperatures up to 450°C, it is possible that chemical attack by the CsCl could be enhanced by carbide precipitation in the grain boundaries due to thermal aging. To determine if thermal aging at 450°C produces potentially harmful microstructural changes, control samples of 316L SS are being aged at temperatures of 400°, 450°, and 500°C for times up to five years. The aged samples are subjected to metallographic examination to determine if microstructural changes have occurred.

The control samples were cut from a rejected WESF inner capsule that did not meet dimensional tolerances, but did meet all other WESF specifications. Each sample was sealed in an argon-filled quartz envelope. The samples are heated in muffle furnaces whose temperatures are maintained within $\pm 2^\circ\text{C}$ of the

control temperatures using solid-state proportioning controllers. The control samples are maintained at temperature for the same times as the thermally aged WESF capsules.

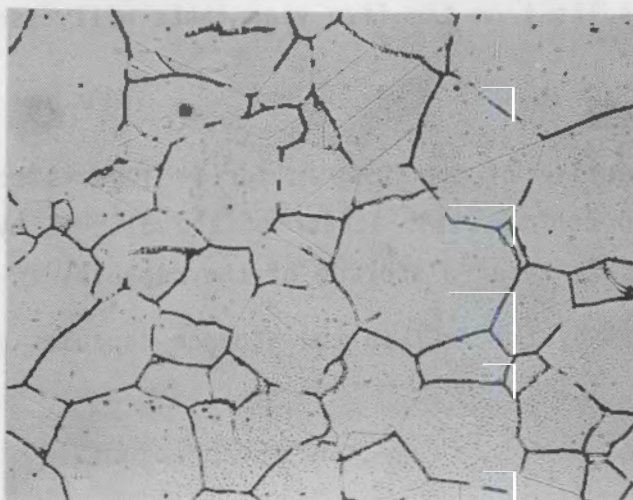
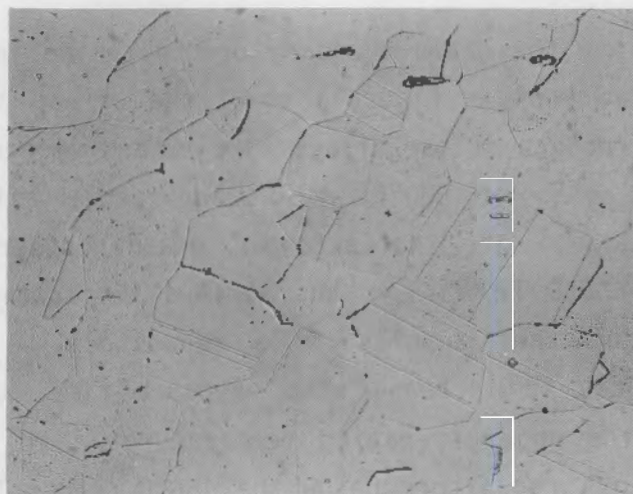
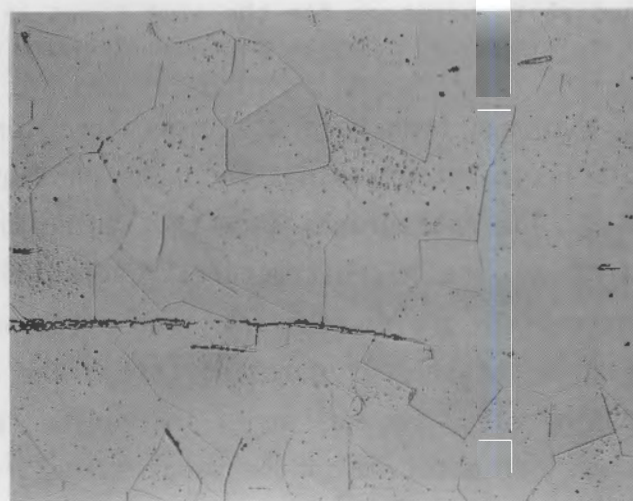
Control samples that had been held at temperature for 28,296 h (3.2 yr) were subjected to metallographic examination. The photomicrographs obtained are shown in Figure 1. The micrographs show that aging the 316L SS at 400°C had very little effect on the microstructure. Aging the alloy at 500°C, however, produced extensive grain boundary precipitation. These results confirm those observed in the shorter duration test. The control sample aged at 450°C for 28,296 h also showed some indication of grain boundary precipitation, which was not observed in the samples aged for shorter times at 450°C.

Figure 2 shows how the grain boundary precipitation in 316L SS increased with exposure time at 500°C. Figure 3 shows the effect of exposure time at 450°C on the microstructure of the alloy. The extensive precipitation resulting from long-time aging at 500°C would undoubtedly affect the corrosion resistance of the alloy. The indication of grain boundary precipitation in the sample aged at 450°C for 3.2 yr has serious implications with regard to long-term disposal of the capsules in a repository. If grain boundary precipitation continues to increase with exposure time at 450°C, it may be necessary to lower the upper operating temperature limit for storage of the WESF CsCl capsules in a repository below the 450°C currently under consideration. Hopefully, results from the five year tests will resolve this issue.

4.3 POOL CELL CAPSULES

A secondary objective of the program for FY 1985 is to obtain sufficient compatibility data to confirm that attack of the 316L SS by the CsCl is not a threat to continued safe interim storage of the capsules in a water basin.

Attack of 316L SS by the CsCl in the storage capsules can be divided into two phases:



40 μ m 500X
FIGURE 1. 316L SS Control Samples Aged for 28,296 h

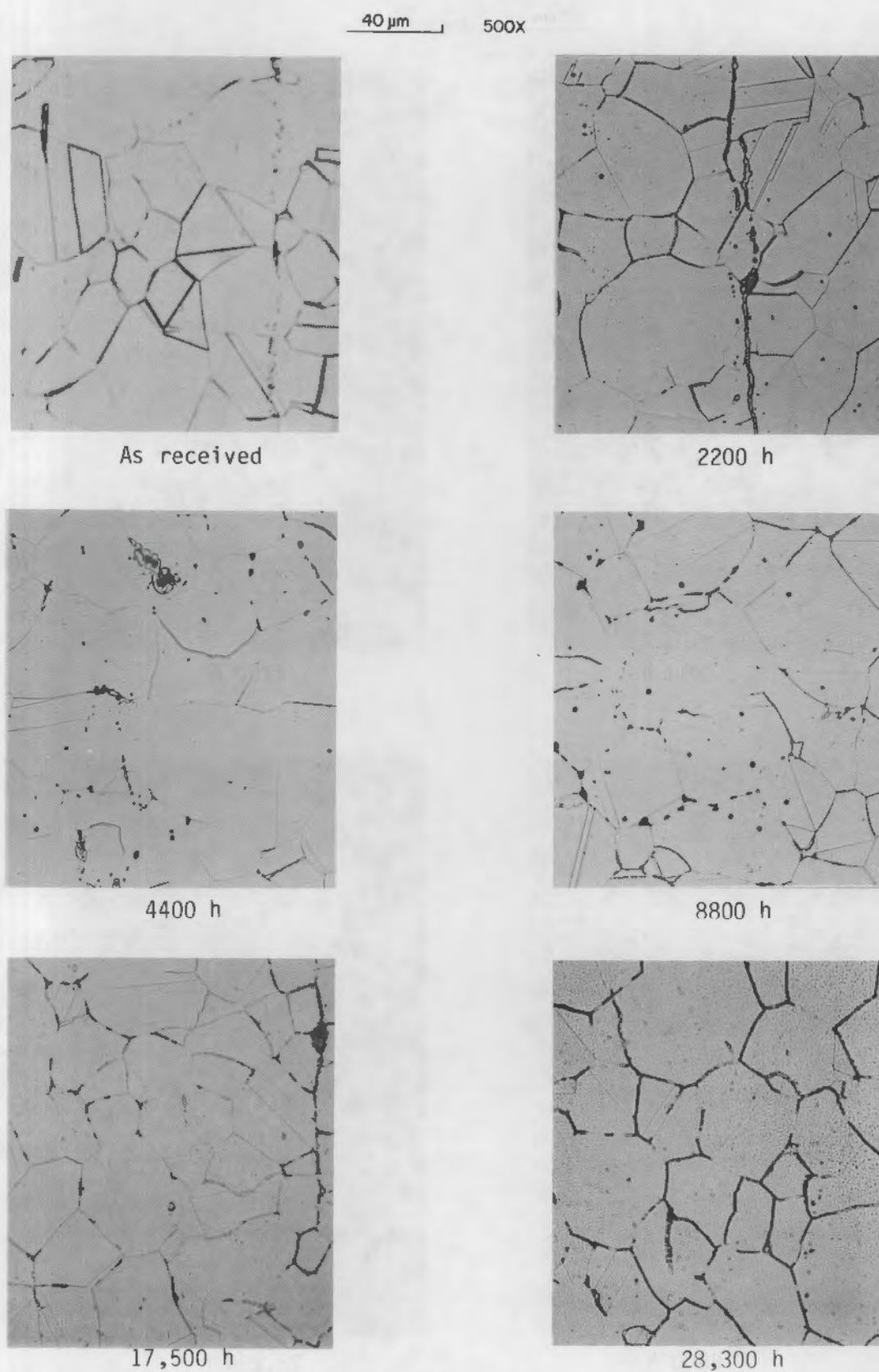


FIGURE 2. Effect of Aging Time at 500°C on the Microstructure of 316L SS

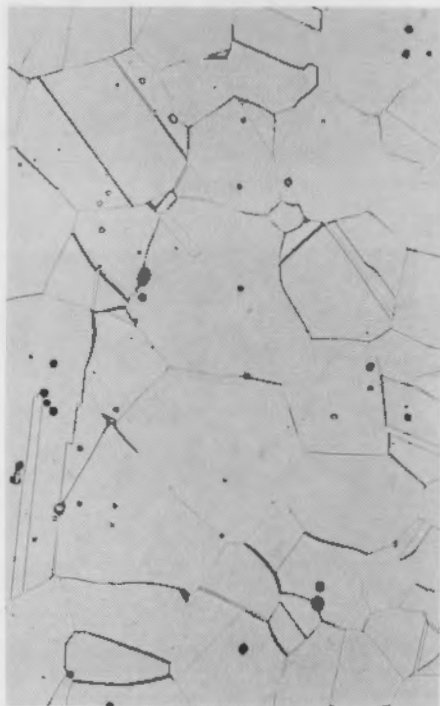
40 μm 500X



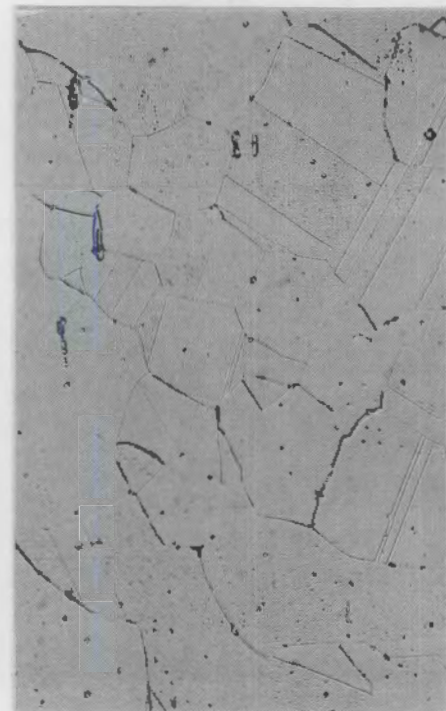
2200 h



4400 h



8800 h



28,296 h

FIGURE 3. Effect of Aging time at 450°C on the Microstructure of 316L SS

- corrosion that occurs during the capsule loading operation when the metal is in contact with molten or hot solid CsCl chloride
- corrosion that occurs during storage in the pool when the metal/CsCl interface temperature is relatively low.

It is necessary to know the extent of corrosion during the loading operation in order to estimate the effect of long-term pool storage on the corrosion.

Two "zero-time" capsules were destructively examined immediately after capsule loading to determine the corrosion resulting from the loading operation. Results obtained from the two "zero-time" capsules were presented in previous reports (Bryan 1983, Bryan 1985) and are summarized below for comparison purposes.

Four samples were taken from each of the "zero-time" capsules for metallographic examination. Corrosion in the capsules was estimated from the photomicrographs obtained, and the results are given in Table 1. Micrographs obtained from the samples are shown in Figure 4. The results indicate that there is little attack of the stainless steel during the loading operation (≤ 0.001 in.). The attack that is observed seems to be somewhat greater in the upper portions of the capsules. The attack appears to involve a general roughening of the metal surface with pitting and limited grain boundary attack at some locations.

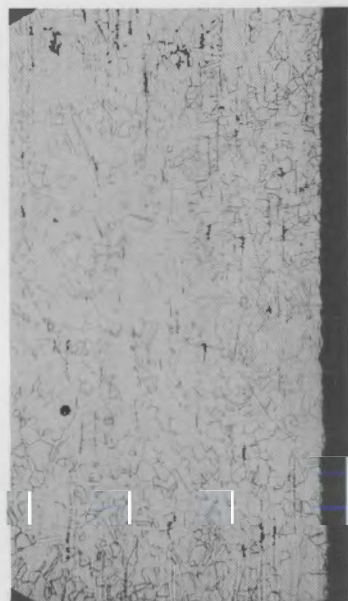
In FY 1985 a total of 18 WESF capsules from the storage pool at WESF were subjected to destructive examination to determine the extent of metal corrosion by the CsCl. The capsules had been stored in the water pool for periods ranging from 0.4 to 8.9 yr. Two storage capsules were also examined in

TABLE 1. Estimates of Metal Attack in the "Zero-Time" Capsules

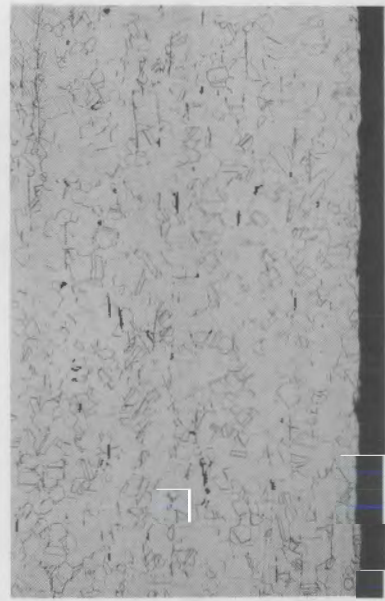
Capsule No.	Depth of Attack, ^(a) μm (mils)			
	Ring No. 1	Ring No. 2	Ring No. 3	Ring No. 4
19459	10 (0.4)	8 (0.3)	25 (1.0)	25 (1.0)
19073F	18 (0.7)	18 (0.7)	25 (1.0)	18 (0.7)

(a) Estimated from photomicrographs.

Capsule No. 19073-F

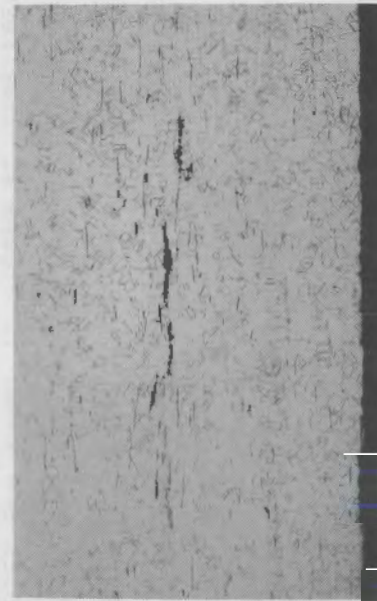


Ring No. 2

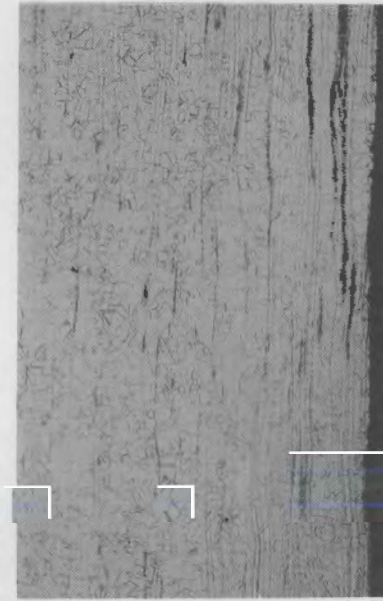


Ring No. 3

Capsule No. 19459



Ring No. 2



Ring No. 3

FIGURE 4. Micrographs of Samples Taken From the Two "Zero-Time" Capsules

200 μ m 100x

FY 1984 (Bryan 1985). With one exception, processing of the 18 capsules was similar to that of the two storage capsules examined in FY 1984 and the two "zero-time" capsules. Only one metallographic sample was taken from each of the 18 inner capsules instead of four samples, as was the case with the capsules examined in FY 1984 and the "zero-time" capsules.

4.3.1 Procedures

Each of the 18 storage capsules examined were placed in the storage pool immediately after filling with CsCl and remained in the pool until removed for destructive examination. After a capsule was removed from the pool, the outer capsule was opened and the inner capsule removed. The inner capsule was opened and any loose CsCl dumped out. The CsCl remaining in the inner capsule was then removed by water washing. After washing, a ring approximately 1-1/2 in. wide was cut from the inner capsule near its midpoint using an abrasive saw. The aforementioned operations were carried out at WESF.

The rings from the 18 capsules were shipped to PNL, where a metallographic specimen was cut from each ring using a diamond saw. The water washing of the inner capsules at WESF reduced the residual radioactivity, principally ^{137}Cs , on the rings to a level where they could be handled directly in an open-faced hood at PNL. The test specimens cut from the rings at PNL were sent to WHC for metallographic examination.

4.3.2 Results

Metal corrosion by the CsCl in the 18 storage capsules was estimated from photomicrographs obtained from the metallographic samples. Wall thickness measurements could not be used to measure the corrosion because initial wall thickness data were not available for the storage capsules.

Estimates of the metal corrosion in the 18 capsules examined in FY 1985, based on the photomicrographs, are given in Table 2. Table 2 also shows the thermal power and ^{137}Cs content of each capsule at the time of loading and its storage time. Because of the limited attack observed in most of the storage capsules, the photomicrographs should provide a reliable measure of the metal corrosion. Micrographs from several of the capsules, showing the different levels and types of attack found, are shown in Figure 5.

TABLE 2. Estimates of Metal Attack in the 18 Pool Cell Capsules Examined

Capsules No.	Capsule Thermal Power, Watts(a)	¹³⁷ Cs Content, kCi(a)	Capsule Storage Period, yr(b)	Depth of Metal Attack, μ m (mils)(c)
C-17	282	58.0	8.9	50 (2.0)
C-88} (d)	214	43.9	8.7	50 (2.0)
C-97}	215	44.1	8.7	30 (1.2)
C-64	266	55.4	8.5	10 (0.4)
C-130} (d)	358	74.5	8.2	80 (3.1)
C-134}	359	74.8	8.2	25 (1.0)
C-206	250	52.1	7.5	15 (0.6)
C-253	259	53.9	7.0	20 (0.8)
C-281} (d)	288	59.9	6.8	30 (1.2)
C-282}	247	51.4	6.8	25 (1.0)
C-413	267	55.7	5.3	40 (1.6)
C-366} (d)	233	48.7	4.6	25 (1.0)
C-368}	234	48.7	4.6	100 (3.9)
C-603	270	56.2	4.1	15 (0.6)
C-809	240	49.9	3.6	25 (1.0)
C-920	240	50.0	3.6	5 (0.2)
C-1777	210	43.7	0.5	25 (1.0)
C-1787	175	36.4	0.4	30 (1.2)

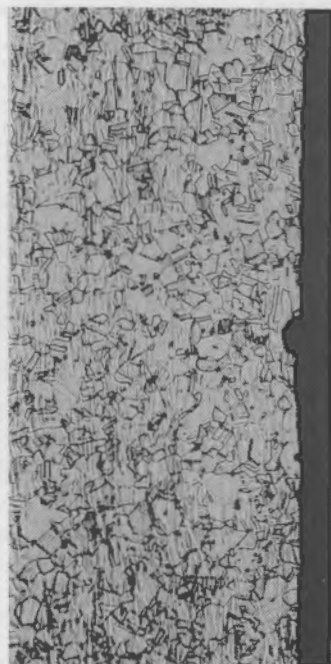
(a) At the time of capsule filling.

(b) Approximate storage period (within 0.1 yr) from the time the capsule was filled until it was sectioned for examination.

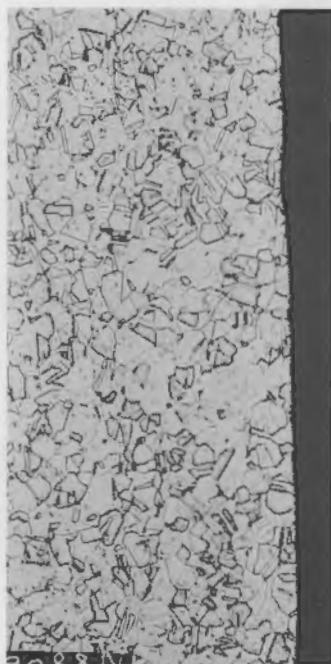
(c) Metal attack was estimated from photomicrographs of the metallographic specimens cut from the midpoint of each inner capsule.

(d) Bracket indicates that the pair of capsules were filled from a given batch of molten CsCl.

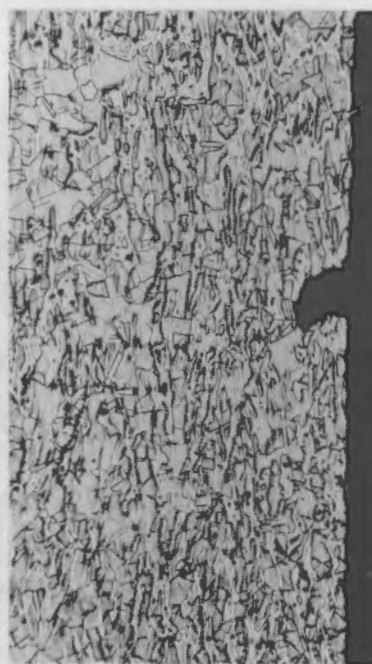
200 μ m 100X



Capsule No. C-1787
Storage Time - 0.4 yr



Capsule No. C-920
Storage Time - 3.6 yr



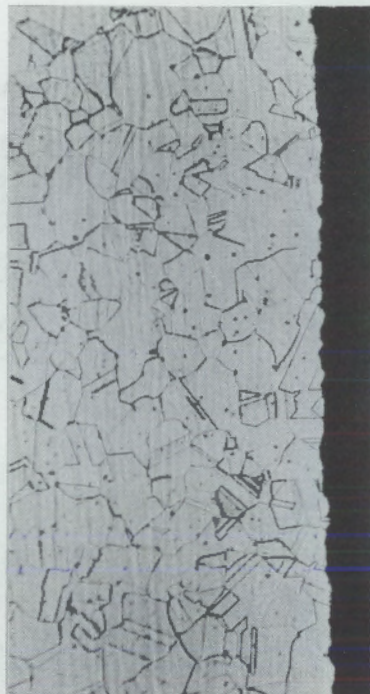
Capsule No. 368
Storage Time - 4.6 yr



Capsule No. C-282
Storage Time 6.6 yr

FIGURE 5. Micrographs of Samples Taken From the Pool Cell Capsules

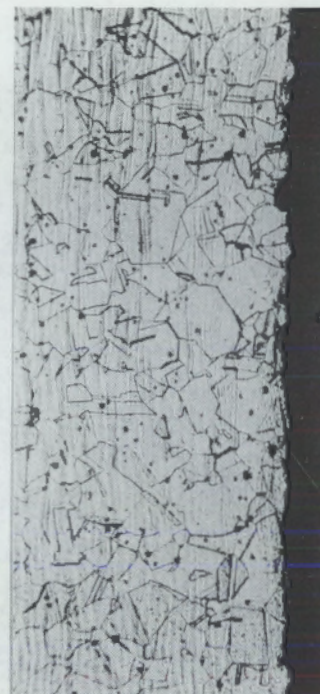
200 μ m 100X



Capsule No. C-206
Storage Time - 7.5 yr



Capsule No. C-130
Storage Time - 8.2 yr



Capsule No. C-97
Storage Time - 8.7 yr



Capsule No. C-17
Storage Time 8.9 yr

FIGURE 5. (contd)

For comparison purposes, results obtained from the two storage capsules examined in FY 1984 are presented in Table 3. Micrographs of samples taken from these capsules are shown in Figure 6. Each of the capsules were sampled at four locations (Bryan 1984). The sample rings taken from the 18 capsules examined in FY 1985 were removed from the canister of the capsule, a location slightly above where the No. 2 rings were cut from the capsules examined in FY 1984 (and the two "zero-time" capsules).

Evaluation of the micrographs from the 20 storage capsules examined in FY 1984 and FY 1985 shows that, on an overall basis, metal corrosion by the chloride is quite low. The corrosion in the capsules appears to involve a general surface attack and pitting. There was little indication of grain boundary attack in most of the capsules. In those capsules where the pitting depth was greatest, the general surface attack appeared to be reduced significantly. The general nature of the pitting appears to be similar to pitting observed in Series 300 stainless steel exposed to aqueous halide solutions.

Based on the limited data available it is difficult to explain the differences in the extent and types of attack found in the storage capsules. In analyzing the data, however, two factors should be recognized:

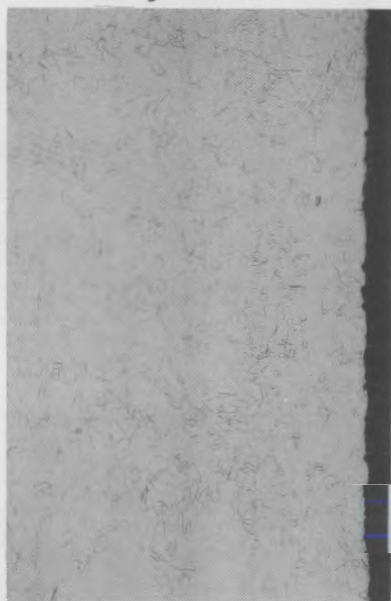
- In compatibility testing it is not uncommon to observe wide variations in attack in replicate tests.

TABLE 3. Estimate of Metal Attack in the Storage Capsules Examined in FY 1984

Capsule No.	Storage Time, yr	Depth of Attack, ^(a) μm (mils)			
		Ring No. 1	Ring No. 2	Ring No. 3	Ring No. 4
C-312	5	16 (0.6)	25 (1.0)	20 (0.8)	18 (0.7)
C-67	8	12 (0.5)	16 (0.6)	14 (0.6)	18 (0.7)

(a) Estimated from photomicrographs.

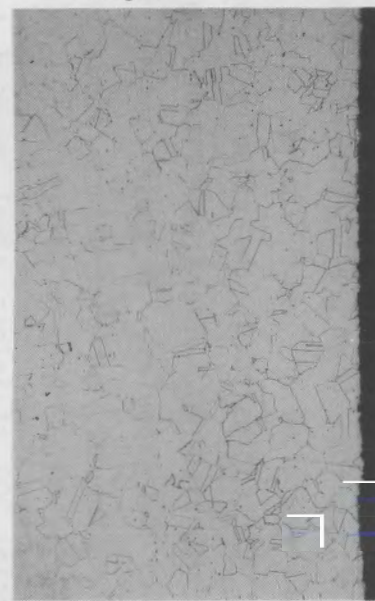
Ring No. 3



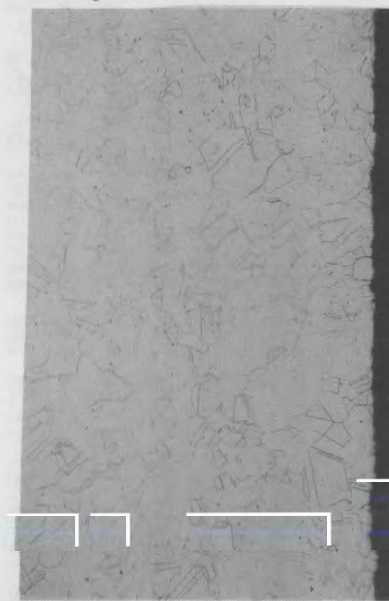
Ring No. 2



Ring No. 3



Ring No. 2



Capsule No. C-312
Storage Time - 5 yr

200 μ m 100x

Capsule No. C-67
Storage Time - 8 yr

FIGURE 6. Micrographs of Samples Taken From the Storage Capsules Examined In FY 1984

- Only one sample, having an area of $\sim 1.6 \text{ cm}^2$, was taken from most of the storage capsules and only four samples from two of the storage capsules and the "zero-time" capsules. Therefore, the metal surface from each capsule that was examined represented only a small fraction of the exposed metal surface ($<1\%$). A more extensive examination of the capsules may have provided more uniform results.

Only slight pitting was observed in the two "zero-time" capsules examined. This could indicate that the pitting corrosion may occur principally during storage; however, only two "zero-time" capsules were evaluated. A more extensive examination of the capsules may have shown the extensive pitting occurred in all capsules including the "zero-time" capsules.

There appears to be little correlation between the depth of the metal corrosion and the storage time in the pool. At least half of the storage capsules examined, some with storage times greater than eight years, exhibited no more corrosion than the "zero-time" capsules. This would indicate that for these capsules, at least, most of the corrosion probably occurred during the loading operation. Attempts to use curve fitting to identify a probable rate equation were largely unsuccessful. For every rate equation evaluated (i.e., linear, logarithmic, parabolic, etc.) the coefficient of determination had a value of less than 0.05 indicating a very poor fit with the experimental data. Again, a more extensive examination of the capsules may have produced more uniform data and permitted identification of the applicable rate equations.

Impurities in the CsCl can have a major effect on the metal corrosion. Most of the storage capsules examined were filled from different batches of cesium chloride, and may, therefore, have contained markedly different types and amounts of impurities. This could help account for the scatter in the corrosion results obtained. The 20 storage capsules examined, however, included four pairs of capsules, where each pair was filled from a given batch of CsCl. The impurity content of each pair of capsules should be very similar. In addition, each pair of capsules were in storage for the same length of time prior to examination. Therefore, theoretically the corrosion in each pair of capsules should be quite similar. This was found to be the case for two pairs of capsules. Each of the other pairs, however, showed

widely different levels of corrosion. In each case one capsule of the pair exhibited pitting that was 3 to 4 times deeper than the pitting in the other capsule. There is no reasonable explanation for such differences unless it reflects a failure to thoroughly examine each capsule.

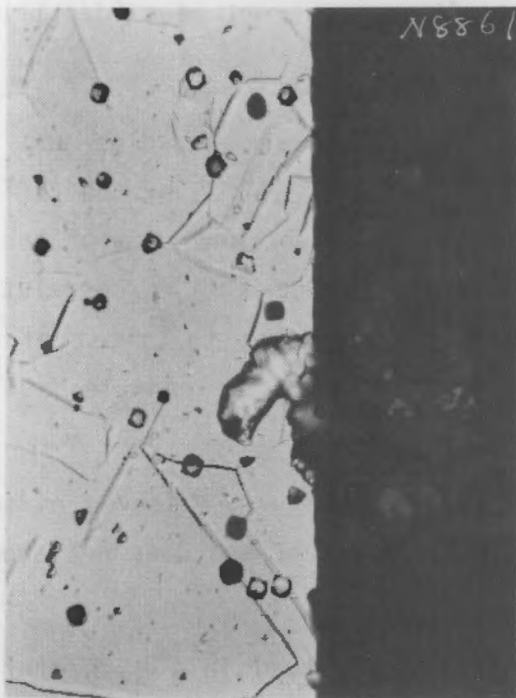
From the above discussion, it is apparent that there is insufficient data to provide a detailed explanation of metal corrosion by the CsCl in the storage capsule. The data indicates, however, that the corrosion in the capsules is quite low [$100\text{ }\mu\text{m}$ (3.9 mils) or less].

Pitting corrosion was observed on the outer surfaces of some of the WESF inner capsules shipped to ORNL for examination. Work carried out at PNL in FY 1984 showed that the pitting could be attributed to problems with the electropolishing operation that was used to decontaminate the inner capsule after filling (Bryan 1985). Specifically, it was determined that the pitting was caused by a nonuniform current distribution at the support rack/capsule contact, which formed localized hot spots. The high temperature caused the phosphoric acid electrolyte to become more concentrated through boiling, and the concentrated boiling phosphoric acid caused a high localized rate of corrosion that produced the pit.

Each sample taken from the 18 inner capsules was also examined for corrosion on the outer surface. Of the 18 samples examined, only four exhibited significant corrosion on the outer surface. In each of the four cases the attack consisted of isolated pitting to depths up to $60\text{ }\mu\text{m}$ (2.4 mil). There was little indication of grain boundary attack or general surface corrosion. Micrographs of the four samples exhibiting the pitting attack are shown in Figure 7. (The dark spherical blotches shown in the micrographs are etchant marks and not part of the metal microstructure.) The nature of the pitting is markedly different from that observed on the inner capsules examined at ONRL. Considering the differences in pit morphology and the pit locations,^(a) it is unlikely that the pitting observed on the storage

The laboratory control of each pair of capsules should be varied. In addition, each pair of capsules were in storage for the same length of time prior to examination. Therefore, theoretically the corrosion

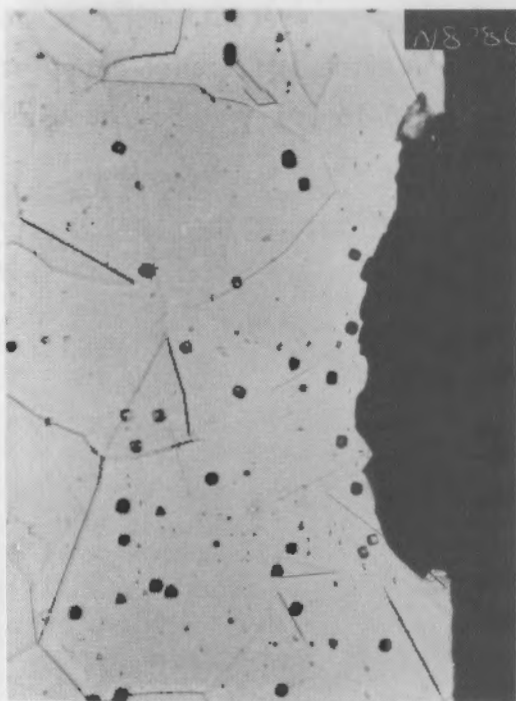
(a) The samples were taken from each inner capsule near its midpoint, which is a substantial distance from any point of support rack/capsule contact.



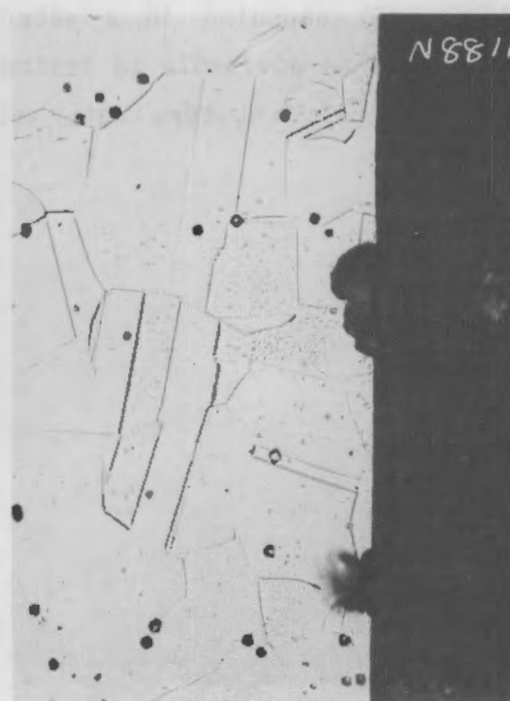
Capsule No. C-17
Storage Time - 8.9 yr



Capsule No. C-64
Storage Time - 8.5 yr



Capsule No. C-282
Storage Time - 6.8 yr



Capsule No. C-940
Storage Time - 3.6 yr

FIGURE 7. Micrographs Showing Pitting Corrosion on the Outer Surface of Samples Taken from the Inner Storage Capsules

capsules can be attributed to the electropolishing operation. To date, however, other possible explanations for the pitting have not been identified.

4.3.3 Conclusions

Evaluation of the corrosion data for the 20 storage capsules and two "zero-time" capsules examined showed that attack of the 316L SS by the CsCl is low. Although there are marked differences in the extent and type of attack observed in the various capsules, there appears to be little correlation between metal attack and storage time. The data indicates that most of the attack observed in the capsules probably occurred during the capsule filling operation.

External corrosion, consisting of isolated pitting, was observed on some of the storage capsules. The attack was limited, however, and does not appear to represent a threat to capsule integrity.

Overall, the internal and external corrosion observed in the 20 storage capsules examined does not appear to seriously affect capsule integrity. Therefore, the corrosion found should not prohibit the long-term safe storage of the WESF CsCl capsules in a water pool. As a precautionary measure, however, it would be advisable to destructively examine additional capsules on a periodic basis if long-term water storage (>10-15 yr) of the capsules is planned.

5.0 REFERENCES

- Bryan, G. H. 1984. Cesium Chloride Compatibility Testing Program Annual Report - Fiscal Year 1983. PNL-4847, Pacific Northwest Laboratory, Richland, Washington.
- Bryan, G. H. and J. R. Divine. 1985. Cesium Chloride Compatibility Testing Program Annual Report - Fiscal Year 1984. PNL-5341, Pacific Northwest Laboratory, Richland, Washington.
- Fullam, H. T. 1982. Cesium Chloride Compatibility Testing Program Annual Report - Fiscal Year 1982. PNL-4556, Pacific Northwest Laboratory, Richland, Washington.

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