MODEL 9975 LIFE EXTENSION PACKAGE 1 – FINAL REPORT

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Model 9975 Life Extension Package 1 – Final Report

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Summary

Life extension package LE1 (9975-03382) was instrumented and subjected to a temperature / humidity environment that bounds KAMS package storage conditions for 92 weeks. During this time, the maximum fiberboard temperature was ~180 °F, and was established by a combination of internal heat (12 watts) and external heat (~142 °F). The relative humidity external to the package was maintained at 80 %RH.

This package was removed from test in November 2010 after several degraded conditions were observed during a periodic examination. These conditions included degraded fiberboard (easily broken, bottom layer stuck to the drum), corrosion of the drum, and separation of the air shield from the upper fiberboard assembly. Several tests and parameters were used to characterize the package components. Results from these tests generally indicate agreement between this full-scale shipping package and small-scale laboratory tests on fiberboard and O-ring samples. These areas of agreement include the rate of fiberboard weight loss, change in fiberboard thermal conductivity, fiberboard compression strength, and O-ring compression set.

In addition, this package provides an example of the extent to which moisture within the fiberboard can redistribute in the presence of a temperature gradient such as might be created by a 12 watt internal heat load. Much of the moisture near the fiberboard ID surface migrated towards the OD surface, but there was not a significant axial moisture gradient during most of the test duration. Only during the last inspection period (i.e. after 92 weeks exposure during the second phase) did enough moisture migrate to the bottom fiberboard layers to cause saturation.

A side effect of moisture migration is the leaching of soluble compounds from the fiberboard. In particular, the corrosion observed on the drum appears related primarily to the leaching and concentration of chlorides. In most locations, this attack appears to be general corrosion, with shallow attack of the drum surface. The primary areas susceptible to corrosion are weld / heat-affected zones. However, one corrosion location not immediately associated with a weld was tested for, and confirmed as having, throughwall penetration.

An increase in the axial gap at the top of the package is also related to the migration of moisture within the fiberboard. As moisture redistributes within the package, a majority of the fiberboard loses moisture (on average) and shrinks axially. In addition, an increased moisture content of the bottom fiberboard layers locally reduces its compression strength, leading to compaction of those layers under the weight of the package internal components. In addition to these moisture-driven phenomena, the fiberboard will shrink due to slow pyrolysis in an elevated temperature / humidity environment. Under the collective influence of these effects, the axial gap in the LE1 package increased from an initial value of 0.58 inch and exceeded the 1 inch (maximum) criterion after approximately 18 weeks conditioning. The axial gap eventually reached 1.86 inches.

Despite the degradation seen in several of the package components (drum, fiberboard, shield and O-rings), the package appears to have retained sufficient integrity to meet the functional requirements for storage in KAMS. This demonstrates a degree of robustness in the package design relative to the storage environment.

Background

This report summarizes information on a 9975 package tested per Task Technical Plan WSRC-TR-2005-00014 [1], which is part of the comprehensive 9975 package surveillance program [2]. This task provides an integrated assessment of the package response to environmental extremes, and demonstrates the extent to which data from small laboratory samples scale up to a full package. The primary goal of this task is to validate aging models currently under development based on lab scale testing of the fiberboard overpack and containment vessel O-rings. A secondary goal is to examine the behavior of the lead shielding under bounding conditions.

Three 9975 packages were modified to provide instrumentation for monitoring package response and performance during environmental aging. Each package has a different environmental exposure history. The first package (LE1, or 9975-03382) began testing October 2007 and was terminated in November 2010. At this point, the fiberboard was darkened and easily damaged. The bottom fiberboard layer was saturated with water, and much of it remained stuck to the drum bottom. The stainless steel air shield had previously detached from the upper fiberboard assembly, creating difficulties in removing the upper fiberboard assembly from the drum. Additional efforts followed to test the mechanical and thermal properties of the fiberboard, and to characterize the containment vessel O-ring seals. This report documents the test data and conclusions related to test package LE1.

Test LE1 was conducted in two phases. In the first phase, the package was maintained at a nominal temperature of 90 °F with no internal heat. The humidity external to the package was held sequentially at 10, 65 and 95%RH while the package was monitored for 6 weeks at each humidity level. The primary focus of LE1 in the second phase was degradation of the fiberboard in an environment with a bulk average temperature of ~160 °F and elevated humidity outside of the drum. In the second phase, the package was in test for 144 weeks, with 92 weeks of that time at the conditioning environment.

The package temperature (142 °F external temperature, ~180 °F maximum fiberboard temperature) is similar to the maximum temperature predicted for a loss of ventilation scenario in storage (137 °F ambient, 192 °F shield maximum temperature with 19W internal heat [3]), and significantly greater than the typical storage ambient temperature of 85 °F or less. The highest recorded ambient temperature in KAMS is 104 °F, although such ambient temperatures are not likely to be sustained for an extended period.

Experimental Method

This life extension package is a 9975 shipping package that was modified to add instrumentation and an internal heat source. An access port for fiberboard sample removal and a view port were added to the drum side wall. Thermocouples provide the temperature at a number of locations throughout the package, including the 3013 payload, PCV, SCV, multiple locations within the fiberboard, and drum surface. A sketch showing thermocouple locations and other modifications is provided in Figure 1.

Package LE1 was placed in an environmental chamber with temperature and humidity control (Figure 2). In the first phase, the package was exposed to three humidity levels (10, 65 and 95 %RH) at a constant temperature of 90 °F, with no internal heat. This evolution was intended to show the degree of moisture infiltration of the package in different humidity levels. For the second phase, an aging environment of 142 °F and 80%RH was maintained in the chamber, and the cartridge heater provided an internal heat source. Initially, the cartridge heater was maintained at 10 watts, but the setpoint was increased to 12 watts after 12 weeks in test (8 weeks at temperature) to better reach the target average fiberboard temperature of ~160 °F.

An additional variable introduced during the second phase was the removal of the package caplugs to increase the movement of moisture into or out of the drum. The caplugs were removed after 5 weeks into the second phase, and were replaced 1 year later.

The PCV and SCV were modified to allow placement of a cartridge heater through the bottom of the containment vessels and into a well in the 3013. The 3013 was welded shut with a surrogate load of steel shot. The cartridge heater conductors and thermocouples attached to the 3013, PCV and SCV exit the package opposite the side where the fiberboard is instrumented, to minimize disruption of the measured fiberboard thermal profile.

The modifications to the PCV and SCV provided open penetrations in the bottom of each vessel. Because of this, both O-rings in each vessel can receive a sensitive helium leak test. Normally, only the outer O-ring is leak-tested with a helium detector, which provides assurance of a leak-tight seal of 1x 10^{-7} std cc air/sec (or 2 x 10^{-7} std cc He/sec). After loading the package, the leak-tightness of both O-rings is typically confirmed at a level of ~1 x 10^{-3} std cc air/sec with a less sensitive rate of pressure rise technique. However, with the modified vessels of LE1, the more sensitive helium leak test can be performed on both O-rings at any time.

Thermocouple data from the package is automatically recorded at preset intervals. Additional data is collected on an occasional basis during periodic examinations. This includes:

- Photographs of the fiberboard position through the view port
- Weight of the entire package
- Weight and moisture content of the removable fiberboard sections
- Relative humidity and temperature at the fiberboard / shield interface
- Visual observation of the package exterior
- Weight and dimensions of the fiberboard assemblies and shield
- Visual observation of other package components

The last 2 of the above data sources require opening the drum lid and removing internal components. These steps are not performed with every examination, so these data are not collected as often as the other items listed.

Results – Periodic Data

There are several metrics which provided evidence of change in the package over time. These include package weight, fiberboard position within the view port, and the temperature profile across the fiberboard. Additional indications of change might be seen in the weight and appearance of the fiberboard, dimensional variation of the lead shield, and from visual observations of the components.

Observations made during the first phase (moisture infiltration check) have been reported previously [4]. They show a general decrease in package weight when exposed to 10 %RH, no significant change in package weight at 65 %RH, and an increase in package weight at 95 %RH. These changes are consistent with a modest degree of moisture exchange with the external environment. No significant degradation was noted during the first phase.

Significant changes in LE1 were observed during the second phase, some of which were reported in Reference 4. These include:

- A strong odor was noted from the removable fiberboard sections after 14 weeks at temperature (17 weeks in test), and continued for the duration of testing.
- The humidity probe inserted through the lower fiberboard assembly failed to fully penetrate through the fiberboard starting after 48 weeks conditioning. This was caused by misalignment between the holes in the drum and fiberboard due to shrinkage / settling of the fiberboard. Humidity data collection continued for 15 additional weeks, but was discontinued in subsequent testing.
- The moisture content of the fiberboard gradually changed throughout the test period. Moisture decreased on the inner surfaces, increased slightly on the outer side surfaces, and increased significantly on the bottom surface (Table 1, Figure 3).
- The appearance of light corrosion was noted on the drum ID surface along the bolting flange stitch welds after 18 weeks at temperature (Figure 4). No significant change in this was noted until the 92 week examination, when the corrosion was much more extensive.
- Corrosion of the shield was heavy after 18 weeks at temperature, and visibly heavier after 39 weeks at temperature. Little change was evident subsequently. (Figures 5, 6)
- After 63 weeks conditioning, the air shield detached from the upper fiberboard assembly.
- After 92 weeks conditioning, much of the bottom fiberboard layer remained stuck to the drum bottom when the fiberboard was removed for inspection.

A plot of package weight over time is shown in Figure 7. Two small sections of fiberboard were cut from the bottom of the lower assembly, and can be removed through a hatch on the drum side. These sections are characterized more often than the drum is opened to inspect the upper or lower fiberboard assembly. Weight and moisture data for these removable sections are shown in Figure 8. Their moisture content is also summarized in Table 1.

Photographs of the removable sections help to track fiberboard changes over time. A sequence of these photographs is shown in Figure 9. The bottom fiberboard layer tends to accumulate the most moisture, as seen in Table 1 and Figure 3. However, the bottom fiberboard layer did not become saturated until the final inspection after 92 weeks conditioning. It was also significantly darkened at this time. At no time was mold observed on the fiberboard, although a strong odor (e.g. rotting vegetation) had developed after 14 weeks in test. Since the entire package exterior was maintained at 142 °F, this elevated temperature may have precluded the growth of mold, although other mechanisms for breaking down fiberboard were active, as evidenced by the odor.

The drum was modified to include a view port on the side. A pattern was marked on the lower fiberboard assembly to be viewed through the view port, and identify the extent to which the fiberboard moved. Typical photographs of this pattern through the view port are shown in Figure 10. The overall fiberboard height was reduced, as indicated by a downward shift in the pattern seen through the viewport. Measurements were made from photographs taken at each inspection interval to estimate the amount of fiberboard movement. These measurements are summarized in Figure 11. Also shown in Figure 11 is the axial gap at the top of the package. As expected, the axial gap increased as the fiberboard height seen through the viewport decreased. The axial gap would have exceeded the 1 inch (maximum) criterion shortly after the 18 week inspection (at which time it averaged 0.994 inch).

Given the relatively low melting temperature of lead (621 ºF), the shield may experience creep at service temperatures. Evidence of this has been seen in life extension package LE2, which was heated to a maximum fiberboard temperature of ~250 °F [5]. Measurements of the LE1 shield were analyzed to investigate this possibility, and are summarized in Table 2.

The corrosion of the lead shield is also of interest. In LE1, moderately heavy corrosion was observed during the first inspection, after 18 weeks in test, and included tan and orange regions. This is in sharp contrast to the corrosion typically seen on a 9975 shield (smooth white surface). After 39 weeks conditioning, the corrosion was mostly white and brown, with a dark blue area near the top (Figure 5). Little subsequent change was noted (Figure 6). Chemical analysis of corrosion product collected from the LE2 package showed that it was generally consistent with the lead carbonate seen on other packages, with elevated levels of chlorine present [5]. Given the similarity in appearance of this shield and the LE1 shield, it is assumed that the corrosion product composition is similar.

Twelve thermocouples monitor the temperature gradient within the fiberboard. The hottest measured temperature within the fiberboard is at the highest thermocouple elevation on the ID surface, and the coolest measured temperature within the fiberboard is at the lowest thermocouple elevation on the OD surface. The temperature vs time data at the highest fiberboard ID and lowest fiberboard OD locations are summarized in Figure 12. It is possible that the fiberboard reached slightly higher temperatures at elevations above the uppermost ID thermocouple. However, the variation among the 4 thermocouples along the ID surface in the central region adjacent to the shield shows a modest gradient (~2 ºF). Therefore, the recorded temperatures are assumed to provide a reasonable approximation of the maximum fiberboard conditions.

Along the OD surface, the four thermocouples indicate a smaller vertical gradient (~1 °F) than along the ID surface. This is consistent with the uniform heating of the package exterior within the environmental chamber. Two of the thermocouples on the fiberboard OD surface failed after ~60 weeks.

Results – Final Inspections

Based in part on the degradation of LE1 fiberboard and the air shield becoming detached from the upper fiberboard assembly, it was decided that the package would not return to test after 92 weeks conditioning. Another key consideration was the appearance of corrosion on the drum interior, as well as local regions on the drum exterior.

Drum Corrosion
On the drum exterior surface, local spots of corrosion were observed on and near the stitch welds around the bottom of the drum (Figure 13). At least 1 of these locations (labeled "A" in Figure 13) was associated with corrosion at the same location on the inside surface. After a light cleaning, both surfaces at this location showed pitting (Figures 14, 15), and throughwall penetration was confirmed by putting water inside the drum and observing moisture on the outside surface.

On the interior drum surface, corrosion was observed in conjunction with most of the fabrication welds. These welds were generally made from the outside, but were still associated with corrosion on the inside surface as a result of the weld heat-affected zone. This observation applies both to the original drum fabrication welds and to the welds made during modification. Typical photos of the corroded areas are shown in Figures 16 - 19. In Figure 16, heavy corrosion is seen around a top flange stitch weld. This is a more advanced stage of the very light corrosion noted after 18 weeks (Figure 4). After cleaning (Figure 17), a general corrosion attack of the surface is observed. Most of the corrosion was dark red - brown to black in color. Some staining from adherent fiberboard was also noted in some areas. In contrast, the corrosion behind the bar code plate attachment welds was white (location "B" in Figure 16). After one of these locations was cleaned, a light general corrosion of the surface was also observed.

When the package was opened for the 92 week inspection, many of the corrosion areas on the inside were observed to be moist, with droplets of a thick liquid running a short distance down the side of the drum. This appearance suggested the possibility of microbiologically influenced corrosion (MIC) activity. Accordingly, Environmental Biotechnology (C. Berry) was contacted and requested to test for microbiological activity. Samples for MIC testing were collected later the same day, but much of the liquid had dried at that point. Testing for MIC proved negative, as summarized in Reference 6:

> "Based on the results from the test kit microbial influenced or initiated corrosion is not indicated for the sampled drum. Viable counts of aerobic bacteria, anaerobic bacteria, sulfate reducing bacteria (SRB), and acid producing bacteria (APB) were determined using a commercially available kit (MICKit™ III Testing, Bioindustrial Technologies, Inc., Georgetown, TX). Swabs were used to swipe the exterior of the drum in areas where discoloration and odors were observed. The swipes were placed in sterile buffer solution and then analyzed using these kits. Serial dilution and aseptic techniques were used to inoculate the test kit. After more than 28 days of incubation at room temperature (i.e., ± 25°C) the kits were visually examined for growth (i.e., turbidity), sulfate reduction (i.e., black precipitate), and acid production (i.e., change in medium color) as instructed by the manufacturer. The results only showed activity for anaerobic bacteria.
>
> "Anaerobic bacteria live on the surface of the drum and are able to grow without oxygen. These concentrations are generally low on solid surfaces, but there may be pockets and crevices of anaerobiosis where these bacteria can grow. These pockets of anaerobiosis have been shown to have an impact on the initiation of MIC (Videla, 1996). The density of

anaerobic bacteria found were quite low, as only one positive results was obtained, and a much larger surface area was sampled, compared to the Kit manufacturers guidance. The estimated density of anaerobic bacteria is less than 1 organism per square centimeter."

Samples of the corrosion product were collected from three areas for analysis in the scanning electron microscope using energy dispersive x-ray spectroscopy. These areas included a stitch weld near the bottom of the drum exterior (location B in Figure 13), a stitch weld near the top of the drum interior (similar to location A in Figure 16), and behind a bar code plate attachment weld on the drum interior (location B in Figure 16). Results are summarized in Table 3.

O-Ring Characterization

The LE1 SCV and PCV were examined visually, received a final leak test, and each vessel was opened to examine the O-rings. No corrosion was observed on either vessel. The leak test on each containment vessel interrogated both O-rings, and the measured leak rate met the acceptance criterion of <2.0 E-7 std cc He/sec. O-ring measurements were taken following several time intervals after the vessels were opened. The first measurements are based on photographs of the O-rings still installed on the cone seal plug taken 7 - 11 minutes after opening. Subsequent measurements were taken with a snap gage after the O-rings were removed, at periods of <30 minutes, 13 days and 30 days after opening. Compression set of each O-ring is calculated based on each of these measurement periods. Results are summarized in Table 4.

Previous tensile testing and DMA analysis of the LE2 PCV inner O-ring [5] showed no significant evidence of aging. Since the LE1 O-rings experienced a lower test temperature, a similar result of no significant degradation was expected, and these tests were not repeated.

Fiberboard Characterization

Samples were removed from the lower fiberboard assembly for thermal conductivity, specific heat capacity and compression testing. During the period between package conditioning and removing the fiberboard test samples, the moisture gradient in the fiberboard diminished significantly, and some overall moisture loss also likely occurred. The moisture content of the bottom layers had dropped from ~100 %WME to ~9 – 10 %WME, while the moisture content of the sides equilibrated to a similar value. Two of the compression test samples from the base of the lower assembly were exposed to ~100 %RH for 1 day prior to testing, to increase the moisture content closer to their condition during package conditioning. The moisture content of each of these 2 samples varied between 16 and 23 %WME. These two samples were tested in the two orientations (one parallel and one perpendicular).

Results of the compression tests are shown in Figures 20 and 21, for the parallel and perpendicular orientations, respectively. Metrics from these tests which provide a basis to draw sample-to-sample comparisons are summarized in Table 5. The buckling strength (applicable only to parallel orientation samples) is the stress at which the fiberboard layers begin to buckle. The area under the stress-strain curve up to 40% strain provides a relative indication of the energy absorption capability of the sample.

Results of the thermal conductivity testing are summarized in Table 6. Samples from the side and bottom regions of the lower assembly were tested at each of three mean temperatures – 77, 122 and 185 °F. No attempt was made to return these samples from the bottom region to their previous elevated moisture level, since elevated moisture interferes with reaching equilibrium during the thermal conductivity test.

Three specific heat capacity samples were removed from the lower fiberboard assembly and tested. At a mean temperature of ~125 °F, the measured specific heat capacity is 1420 J/kg-K. This value is the average of 9 trials, conducted in accordance with ASTM C351. The specific heat capacity measured on other 9975 packages that were destructively examined ranged from 1300 – 1875 J/kg-K. This result for aged LE1 fiberboard material is consistent with that from the other packages.

Discussion

The LE1 environment is considered bounding to the typical package storage conditions in KAMS. The humidity outside the package (80%RH) combined with the elevated temperature maintains a much higher moisture content in the air than in storage. On the other hand, the steady decrease in package weight through the second phase of testing and the moderate moisture levels throughout the period indicate there was not a significant ingress of moisture into the package.

The fiberboard temperature during the second phase of testing ranged from 142 °F at the exterior surface to ~180 °F in the interior, with an average of ~160 °F. This range of environments provides an opportunity to compare the fiberboard behavior to several environments used for laboratory-scale testing of fiberboard. These include 160 °F 50 %RH, 185 °F 30%RH, and 185 °F dry oven. Although the package temperature (142 °F external temperature, ~160 °F bulk fiberboard temperature) is similar to that predicted for a loss of ventilation scenario in KAMS, it is significantly above the typical facility ambient temperature of 85 °F or less.

Testing of package LE1 was terminated based on several observations that indicated the package had degraded to a point that compromised the validity of further testing. Specifically,

- The air shield had detached from the upper fiberboard assembly during the previous inspection.
- Significant corrosion had initiated on essentially all the drum interior surfaces associated with welds and weld heat-affected zones, and on the exterior surface around some of the bottom stitch welds.
- Much of the bottom fiberboard layer remained stuck to the drum bottom when the lower assembly was removed. Other regions of the lower assembly were fragile and easily damaged from normal handling.

In spite of these shortcomings, it is noted that the package appears to have remained capable of fulfilling its safety functions in a storage environment. For example,

- The large majority of fiberboard remained present (2.8 kg total weight loss vs 35 kg total fiberboard weight) to perform a criticality control function.
- The fiberboard thermal conductivity changed at a modest rate of ~2.5 %/year. Both thermal conductivity and specific heat capacity at the end of life fall within a range consistent with undegraded material.
- The energy absorption capability of the fiberboard, represented by the area under the compression stress-strain curve up to a strain of 40%, remained at 14 psi or higher. The

package has been shown to survive a dual forklift impact with the fiberboard strength reduced to 20% of nominal [7], which corresponds to an area under the stress-strain curve up to 40% of 11 psi.

- Despite at least one point of corrosion penetrating through the drum wall, the bulk of the drum remained intact. The majority of the drum surface was not affected by corrosion, and the majority of affected areas examined sustained only a very shallow corrosion attack.
- The containment vessels experienced no corrosion or other degradation, and the O-rings remained leak-tight, ensuring that the package would still perform its containment function.

This test package therefore demonstrates that the 9975 package is robust and can perform its required functions in spite of the challenging bounding environment it was exposed to for 92 weeks. It is possible that some of the observed degradation was aggravated by being removed from test for 40 weeks following 63 weeks exposure (due to the environmental chamber being unavailable). During this time, the thermal gradient in the fiberboard would have quickly disappeared, and the moisture gradient within the fiberboard was significantly reduced. Upon restarting the test, the moisture gradient was re-established. This created additional cycles of significant moisture migration along with the potential for leaching of chlorides and other compounds from the fiberboard. Therefore, the observed degree of drum corrosion may overestimate the rate of attack likely to occur under storage conditions, and cycling of these LE1 conditions likely adds a layer of conservatism to the test environment.

With an elevated external temperature of 142 °F, the moisture within the fiberboard was likely more mobile than it would be under normal KAMS storage conditions with an ambient temperature typically 85 °F or less. At the higher temperature, a greater fraction of the free moisture will be in the vapor phase, making it more mobile within the fiberboard. This would likely accelerate and may exaggerate the moisture gradient that would otherwise develop under storage conditions.

Thermal Conductivity Changes for Package LE1 vs Laboratory Data

Package LE1 data were examined for evidence of change in fiberboard thermal conductivity during the testing. The fiberboard sidewall region has a relatively constant thermal response among the thermocouples on the fiberboard ID and OD surfaces. However, the two upper thermocouples on the OD surface failed during the test, so the lower half of this sidewall region will be examined with regards to its thermal conductivity. Since some heat is lost through the top and bottom of the package, the heat conducting through this side region will be less than 12 watts, but it will be assumed constant over time. Heat conduction in this region is in the radial orientation.

Thermal gradient information was examined at several discrete times during periods of steady state operation indicated by the vertical arrows in Figure 22. The thermal conductivity will vary with changes in the temperature gradient and fiberboard thickness, as described by:

$$q/A = k * \Delta T / t$$

where, q/A = heat flux (assumed constant)
k = thermal conductivity

ΔT = radial temperature gradient
t = lower assembly radial thickness (linear interpolation based on available inspection data)

At each of the vertical arrows in Figure 22, the radial temperature gradient and lower assembly thickness are combined per this relationship to get a value proportional to the thermal conductivity (see Table 7). Any change in this value over time is proportional to the change in actual thermal conductivity. (Since the actual heat flux through this region of fiberboard is unknown, the actual thermal conductivity cannot be calculated directly.)

The value at the first arrow in Figure 22 (after 120 days at temperature) was taken as a reference point, and the values for subsequent data points show the change in thermal conductivity relative to this first data point. These results are listed in Table 7 and plotted in Figure 23, along with normalized laboratory data for samples conditioned at 160 °F 50%RH (sample MSC-4R), 185 °F 30%RH (sample 2234-3R) and 185 ºF dry oven (sample TC3R) and tested at 185 ºF. In this presentation, the relative thermal conductivity values for LE1 show a decrease with time at a rate of ~2.5 %/year, and fall within the range of trends for the laboratory samples. None of the laboratory sample trends show a significant change in thermal conductivity over time (less than 6% decrease over 1.8 years), indicating that actual storage conditions are relatively benign with regards to thermal conductivity performance. Calculations have shown that the 9975 package will perform its thermal insulation function (for normal operation and postulated fire scenario in KAMS) even with 20% reduction in fiberboard thermal conductivity [8]

Fiberboard Physical Changes vs Laboratory Data

Variation in the total weight of the package is summarized in Figure 7. Variation in the weight of the removable fiberboard sections is shown in Figure 8. The minor changes in weight of the removable sections trend well with the changes in moisture content of those sections, indicating relatively little actual degradation. It is assumed that the change in package weight is associated with the fiberboard alone (ignoring effects of weight gain from the shield and drum corrosion). It is further assumed that there was no significant moisture loss from the package. This assumption is based on the observation that the fiberboard moisture content and gradient remained approximately constant throughout most of the testing.

The total package weight decreased at an average rate of 0.0025% /day. The package initially contained 35.311 kg of fiberboard (based on the initial weight of both fiberboard assemblies and removable sections, minus the nominal weight of bearing plates and air shield). Accordingly, the overall average rate of fiberboard weight loss is 0.013 %/day, or 4.7 %/yr. Similarly, using the data from Figure 8, the average fiberboard weight loss rate for the small (upper) removable fiberboard section is 4.1 %/yr, and for the large (lower) removable fiberboard section is 3.3 %/yr (prior to the last inspection which saw a significant moisture (and weight) increase).

The average fiberboard temperature throughout much of the package is close to 160 ºF, and a moderate amount of moisture remains in much of the fiberboard. The average rate of fiberboard weight loss is compared to that from several laboratory test environments (including 160 °F 50%RH) in Table 8. Comparable rates of weight loss are seen in laboratory environments of 160 °F 50%RH and 185 °F 30%RH. Within dry laboratory environments, similar rates of weight loss are not seen until the temperature is as high as 215 °F.

The fiberboard dimensions changed throughout the second phase of testing. The overall height of the fiberboard decreased steadily, as indicated by the increasing axial gap (Figure 11). The rate of height reduction averaged 0.7 inch/year, or about 2% of the total fiberboard height per year. At the same time, the fiberboard thickness was also decreasing, but at a lower rate. From Table 7, we observe the radial thickness decreasing at 0.030 inch/year, or 0.6% of the radial thickness per year.

Similar dimensional changes are seen in the laboratory test environments of 160 °F 50%RH and 185 °F 30%RH.. The ranges of behavior for these two environments overlap, with a total range of height change of 1.3 to 4.5 %/year, and a total range in width change of 0.2 to 1.7 %/year. These rates encompass those observed in LE1. Note that the change in fiberboard height directly impacts the axial gap, while the effect of a change in fiberboard radial thickness will be divided between the fiberboard / drum gap and the fiberboard / shield gap.

Fiberboard Strength Changes vs Laboratory Data

Some variation is seen in the compressive strength of the LE1 fiberboard samples for both test orientations (Figures 20, 21). These figures also show the stress-strain curves for typical un-aged material. Comparative metrics for the compression tests are summarized and compared to laboratory samples in Table 5.

The parallel orientation is relevant to a side impact scenario, such as a forklift impact. For this orientation, the LE1 base sample had higher buckling strength and energy absorption than samples from the side with similar moisture content. This could result from less degradation to the lower fiberboard layers or from these layers coincidentally being stronger to begin with. The slight decrease in weight loss rate for the removable fiberboard sections noted above compared to the entire assembly suggests the lower layers were exposed to a slightly less aggressive environment, although variation in properties throughout a fiberboard assembly has also been observed. For the base sample that was re-conditioned to a higher moisture content prior to testing, both the buckling strength and energy absorption decreased significantly.

In comparison to the data from LE1 samples, compression testing has also been performed on samples conditioned at 185 °F 30%RH for up to 98 weeks. A stress-strain curve for this condition is also shown in Figure 20. The metrics for this case are similar to those for the LE1 package (Table 5). Comparison to test data for un-aged material shows that the LE1 fiberboard experienced a significant reduction in strength and energy absorption. It is noted, however, that the LE1 sample metrics show a minimum area under the stress-strain curve up to 40% of 14 psi. Separate analysis has shown that the package will meet energy absorption requirements during a forklift impact event even with the area under the stress-strain curve up to 40% reduced to 11 psi [7]. Therefore, it is expected that the condition of the fiberboard in LE1 would still meet the energy absorption requirement for storage.

Samples tested with the load applied perpendicular to the fiberboard layers are compared with un-aged material in Figure 21 and Table 5. Similar to the parallel orientation data, the LE1 fiberboard is weakened relative to un-aged material, but should not compromise the mechanical property requirements for storage.

Potential for Shield Creep
Dimensional measurements of the lead shield have been evaluated to determine whether creep of the lead has occurred during testing. This possibility is suggested by comparable data from Life Extension package 2 (9975-04162) [9], although the lower conditioning temperature of LE1 should reduce the magnitude of the creep rate.

The data in Table 2 indicate that no significant creep of the LE1 shield has occurred. If creep had occurred, the shield thickness at the top would tend to decrease while the thickness at the bottom would increase. Any such change is partially masked by the formation of a corrosion layer on the shield, but this layer is relatively uniform in thickness. The changes in shield thickness over time are generally positive and of similar magnitude at both the top and bottom. Therefore, these changes are likely due to corrosion alone, with no significant contribution from creep.

LE1 O-Rings
The O-rings in LE1 were generally at a temperature of ~181 ºF (PCV) or ~172 ºF (SCV), with a total time at temperature of ~650 days. This is lower than the calculated maximum O-ring temperatures in KAMS (198 °F maximum during normal operation based on 137 °F ambient temperature and 19 watts internal heat load) [3], but higher than expected in storage for a more typical ambient temperature of 85 °F.

The compression set of each O-ring measured initially upon opening ranged from 36 to 56%. These values are based on measurements taken from photographs of the O-rings still installed on the cone seal plug. Thickness measurements were taken with a snap gage approximately 20 minutes later, giving generally smaller, but comparable, values. This illustrates the validity of measurements from photographs, which can provide an opportunity to obtain data sooner after opening a vessel. After 30 days, the compression set values had decreased to a range of 11 to 22%.

Data from laboratory testing of O-rings [10] indicates that the compression stress relaxation (CSR) behavior for a similar exposure (175 ºF for 650 days) would be ~35% loss of sealing force. While compression set and compression stress relaxation do not necessarily change at the same rate for a given environment, they might both be expected to change in the presence of a degrading environment. The initial LE1 O-ring compression set was greater than the CSR value for a comparable exposure, while the 30 day compression set values were lower. This same relative trend was observed with the LE2 O-rings [5].

Conclusions

Life extension package LE1 has been instrumented and exposed to a bounding storage condition to help identify the extent to which laboratory test results for fiberboard, O-rings and other components apply to a full-scale package. The following forms of degradation have been seen in this package:

- Corrosion of the drum in the vicinity of welds and weld heat-affected zones likely resulted from the leaching of chlorides from the fiberboard. A moisture gradient formed in the fiberboard, driven by the internal heat load (12 watts in this case). This provided a driving force for leaching soluble compounds from the fiberboard. This moisture gradient may have also been enhanced by the elevated external temperature.

- In the bounding environment (elevated temperature and humidity), the fiberboard loses mass (due to pyrolysis) and shrinks. Shrinkage is greatest in the axial direction. This behavior is consistent with laboratory samples for similar environments.
- The fiberboard compression strength was degraded and shows consistency with laboratory samples aged under similar conditions. The elevated moisture in the bottom fiberboard layers further reduced the compression strength. Some of this strength loss is recovered as the moisture level is reduced. A similar partial recovery is seen in laboratory samples.
- Three phenomena can contribute to an increase in the axial gap at the top of the package:
    - Loss of fiberboard due to pyrolysis,
    - Compaction of the bottom fiberboard layers due to reduced compression strength in the presence of increased moisture levels, and
    - Shrinkage of fiberboard above the bottom layers due to a net decrease in moisture content.

  All three of these phenomena were likely active in this package, and contributed to an axial gap that eventually reached 1.86 inches. The initial 0.58 inch axial gap reached the 1 inch (maximum) criterion after approximately 18 weeks conditioning.
- The fiberboard thermal conductivity decreases over time at elevated temperature. Estimates of the change in thermal conductivity (in the radial direction) are consistent with changes seen in laboratory samples for similar environments.
- The lead shield experienced uniform corrosion, with the morphology differing from that typically seen in 9975 packages. However, the corrosion product appears similar to that observed on the LE2 package, which was identified as consistent with the lead carbonate observed previously plus chlorine.
- Based on laboratory test data on O-rings to date, failure of the O-rings is not expected for the conditions experienced by LE1. The O-rings in the LE1 containment vessels remained leak-tight. Upon opening the LE1 containment vessels, the compression set measured initially ranged from 36 to 56%.

Despite the many forms of degradation observed to the LE1 package components (drum, fiberboard, shield and O-rings), the package appears to have remained capable of performing the functional requirements for storage. The fiberboard retained sufficient mass and compression strength to fulfill its criticality control and impact energy absorption functions. Fiberboard thermal conductivity decreased at a rate of ~2.5 %/year. Recent analysis has shown that significantly greater changes in thermal conductivity can be sustained without compromising the thermal insulation capability to survive a fire scenario in storage. The containment vessels were undegraded, and the O-rings remained leak-tight to perform their containment function. This test package therefore demonstrates a degree of robustness of the 9975 package design relative to the KAMS storage environment.

References

[1] WSRC-TR-2005-00014, Rev. 2, "Task Technical and Quality Assurance Plan for Full-Scale Testing of Model 9975 Packages to Support Life Extension Model Development (U)", June 2008.

[2] WSRC-TR-2001-0286, Rev. 2, “SRS Surveillance Program for Storage of Pu Material in KAMS”.

[3] M-CLC-K-00727, “Thermal Model Study for the 9975 Package in KAMS During Facility Fire”, N. K. Gupta, June 11, 2008.

[4] SRNL-TR-2009-00439, Rev. 1, “First Interim Status Report: Model 9975 Life Extension Package Testing”, W. L. Daugherty and T. M. Stefek, January 2010

[5] SRNL-STI-2010-00185, “Model 9975 Life Extension Package 2 – Final Report”, W. L. Daugherty, April 2010

[6] “Re: Status of MIC Samples”, electronic mail message from Topher Berry to William Daugherty, January 20, 2011

[7] M-CLC-K-00657, Rev. 1, “Structural Analysis of 9975 Package Subjected to Two Forklift Impact”, T. Wu, December 16, 2005.

[8] M-CLC-K-00747, “Thermal Model Study of the 9975 Package with Moisture Degraded Fiberboard in KAMS Facility”, N. K. Gupta, May 18, 2010.

[9] SRNL-STI-2010-00185, “Model 9975 Life Extension Package 2 – Final Report”, W. L. Daugherty, April 2010.

[10] SRNS-TR-2009-00259, “Lifetime Prediction for Model 9975 O-Rings in KAMS”, E. N. Hoffman and T. E. Skidmore, November 2009.

Table 1. Moisture content of removable fiberboard sections for LE1 2nd phase

Time at Temp (days)	Moisture Content (%WME)		
	Near bottom of lower section	Near top of upper section	
0	NA	NA	
50	18.7	11.2	Caplugs removed at 30 days
92	20.3	16.4	Increase internal heat from 10 to 12W at 56 days
121	22.1	15.2	
157	26.5	17.0	
267	17.7	11.8	
267	13.0	8.8	Package out of chamber for 9 days
311	23.6	13.3	Caplugs replaced at 304 days
331	21.2	14.5	
360	31.3	16.0	
393	29.2	12.5	
446	26.3	13.8	
476	22.0	13.4	
544	21.9	13.8	
650	100	15.7	

Table 2. Dimensional measurements of LE1 shield to investigate the potential for lead creep.

Conditioning at Time of Measurement	Avg Radial Thickness at Bottom of Shield (inch)	Avg Radial Thickness at Top of Shield (inch)	Difference (Bottom – Top) (inch)
Baseline	0.559	0.541	-0.018
After 39 wks	0.572	0.556	-0.016
After 56 wks	0.570	0.561	-0.009
After 92 wks	0.586	0.560	-0.026

Table 3. Summary of SEM analysis of drum corrosion product

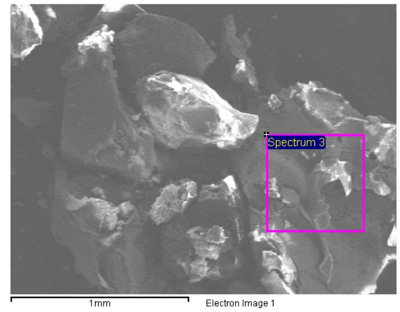
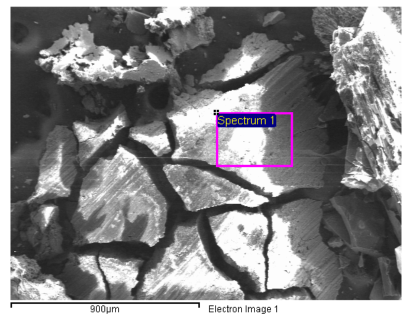
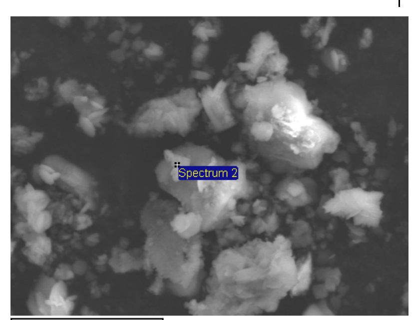
Location	General Results	SEM Results for Typical Particle
 Drum OD surface at bottom stitch weld	8 particles checked 7 particles have some or all stainless steel elements (Fe, Cr, Ni, Mo) plus 8 – 24 wt% Cl 1 particle is "dirt" (Na, Al, Si, Cl, K, Ca, F, O)	52.6 wt% Fe 10.0 wt% Cr 2.5 wt% Ni **12.4 wt% Cl** 0.5 wt% Si 0.4 wt% S 21.6 wt% O 
 Drum ID surface at top flange stitch welds and vertical seam weld	7 particles checked: All particles have some or all stainless steel elements (Fe, Cr, Ni, Mn) plus O 4 particles have 1 - 3 wt% Cl Some have significant amounts of Si and/or Al	43.7 wt% Fe 11.8 wt% Cr **3.0 wt% Cl** 7.6 wt% Al 4.4 wt% Si 29.6 wt% O 
 Drum ID surface behind bar code plate attachment weld	8 locations on 5 particles checked: All locations have some or all stainless steel elements (Fe, Cr, Ni, Mn) plus O 4 locations on 4 different particles have 0.5 – 0.9 wt% Cl. 3 locations on 2 different particles have 1 – 2 wt% Cu.	20.2 wt% Fe 0.6 wt% Cr 19.0 wt% Ni **0.9 wt% Cl** 16.7 wt% Al 4.5 wt% Si 4.1 wt% Na 1.3 wt% Cu 29.6 wt% O 

Table 4. Compression set data for LE1 O-rings after 92 weeks conditioning

	O-Ring Compression Set			
Interval between opening vessel and O-Ring measurements	PCV Inner O-Ring	PCV Outer O-Ring	SCV Inner O-Ring	SCV Outer O-Ring
7 - 11 minutes	36%	43%	44%	56%
24-28 minutes	42%	34%	44%	45%
13 days	22%	12%	23%	10%
30 days	21%	11%	22%	14%

Table 5. Comparative metrics from fiberboard compression tests

Sample	Buckling Strength (psi)	Area under Stress-Strain Curve to 40% Strain (psi)	Sample	Area under Stress-Strain Curve to 40% Strain (psi)	Comment
Parallel Orientation			**Perpendicular Orientation**		
LE1 Side 1	92	15	LE1 Side 3	24	9 – 10 %WME moisture content at time of test
LE1 Side 2	83	20	LE1 Side 4	24	
LE1 Base 1	140	24	LE1 Base 3	37	9 – 10 %WME moisture content at time of test
LE1 Base 2	55	14	LE1 Base 4	17	Elevated moisture content (~18 – 20 %WME) re-established pre-test
Lab Sample Data for Comparison	98 - 212	10 - 25	Lab Sample Data for Comparison	NA	Conditioned 98 wks at 185°F 30%RH
	152 - 357	36 - 78		25 - 61	Non-aged material

Table 6. Thermal conductivity data for LE1 fiberboard lower assembly

Sample Location & Orientation	Thermal Conductivity (W/m-K) for a mean temperature of:		
	25°C (77°F)	50°C (122°F)	85°C (185°F)
Upper side wall, axial	0.0599	0.0631	(0.0594) *
Upper side wall, radial	0.0970	(0.0961) *	0.0976
Bottom layers, axial	0.0684	0.0694	(0.0582) *

* These three results are inconsistent with the remaining data, in that the thermal conductivity typically increases as the temperature increases. This anomaly is likely due to the lack of an equilibrium moisture gradient in the samples during testing.

Table 7. LE1 data used to estimate changes in fiberboard radial thermal conductivity

Conditioning Time (days)	Avg Fiberboard ID Temperature (ºF)	Avg Fiberboard Radial Temp. Gradient *, ΔT (ºF)	Fiberboard Radial Thickness, t (inch)	ΔT / t (ºF/inch)	Normalized Thermal Conductivity, 5.695 / (ΔT / t)
120	168.9	27.49	4.827	5.695	1
255	168.6	27.53	4.819	5.713	0.997
357	168.4	27.26	4.794	5.686	1.002
567	168.7	27.72	4.786	5.792	0.984
636	168.7	28.24	4.784	5.903	0.965

* The radial temperature gradient is based on the two lower thermocouple locations only, since the upper two thermocouples on the OD surface failed during the test.

Table 8. Comparison of fiberboard weight loss rate for package LE1 and several laboratory environments

Environment	Period of Exposure	Range of observed weight loss rate
LE1: ~160F average temperature, some moisture present	650 days	4.7 %/yr all fiberboard
	650 days	4.1 %/yr small removable section
	544 days	3.3 %/yr large removable section
160 °F 50%RH	170 days	3.1 – 5.8 %/yr
185 °F 30%RH	615 days	3.6 – 5.2 %/yr
185 °F dry oven	1422 days	1.0 – 1.3 %/yr
215 °F dry oven	1489 days	3.0 – 3.6 %/yr

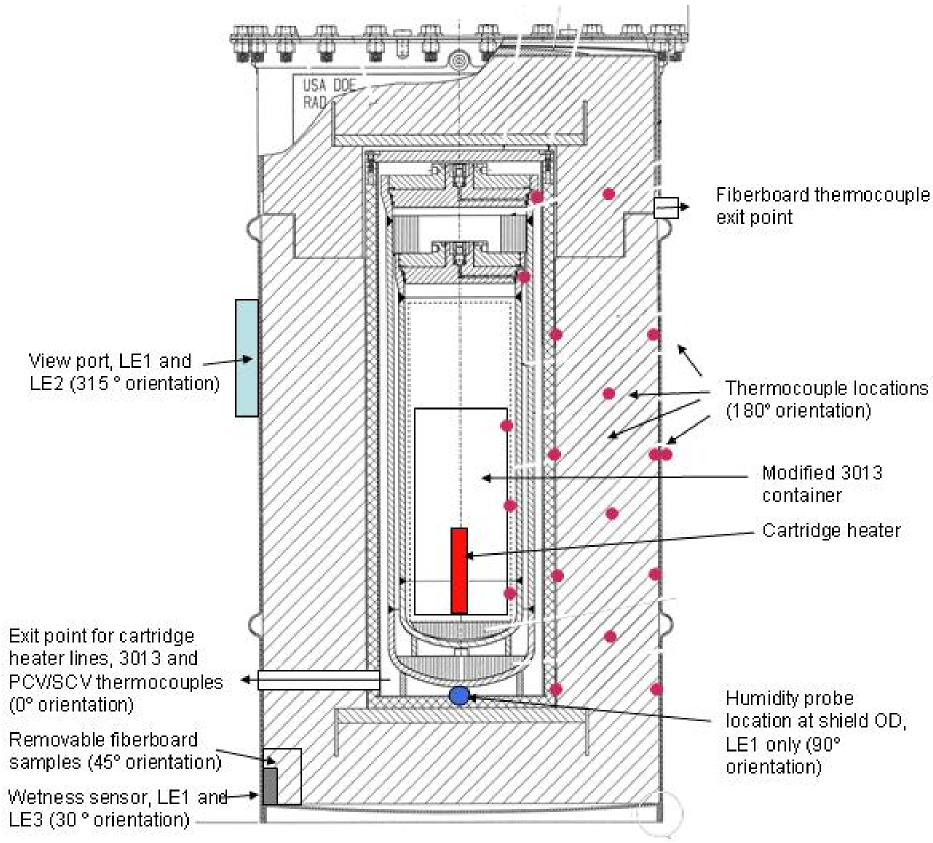
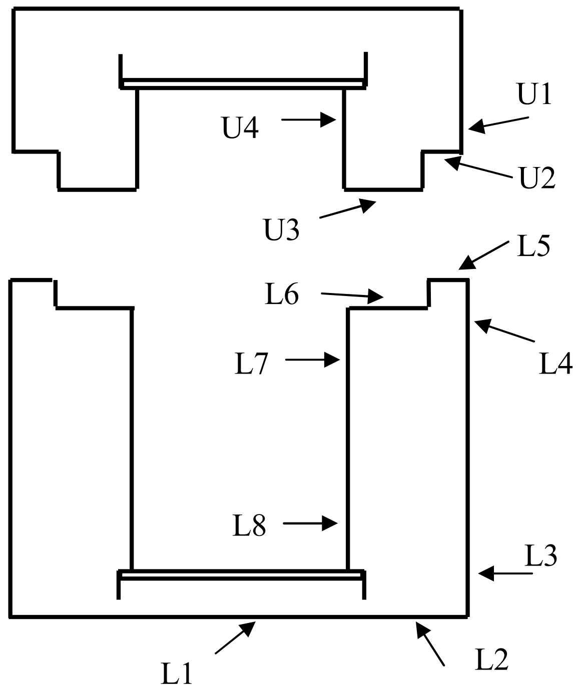


Figure 1. Cross section of 9975 package, showing added instrumentation and examination features.

Figure 2. Configuration of LE1 in environmental chamber



Location	Avg. Fiberboard Moisture (%WME) after:		
	0 wks	39 wks	92 wks
Upper assembly			
U1	12.8	11.2	16.1
U2	14.2	11.6	13.1
U3	13.8	7.2	< 6
U4	13.3	< 6	< 6
Lower assembly			
L1	NA	43.5	100
L2	NA	33.4	44.2
L3	NA	18.4	20.3
L4	NA	13.8	18.5
L5	11.4	10.2	14.5
L6	11.2	6.4	7.0
L7	11.6	< 6	< 6
L8	11.8	< 6	< 6

Figure 3. Summary of fiberboard moisture content variation during LE1 2^{nd} phase.

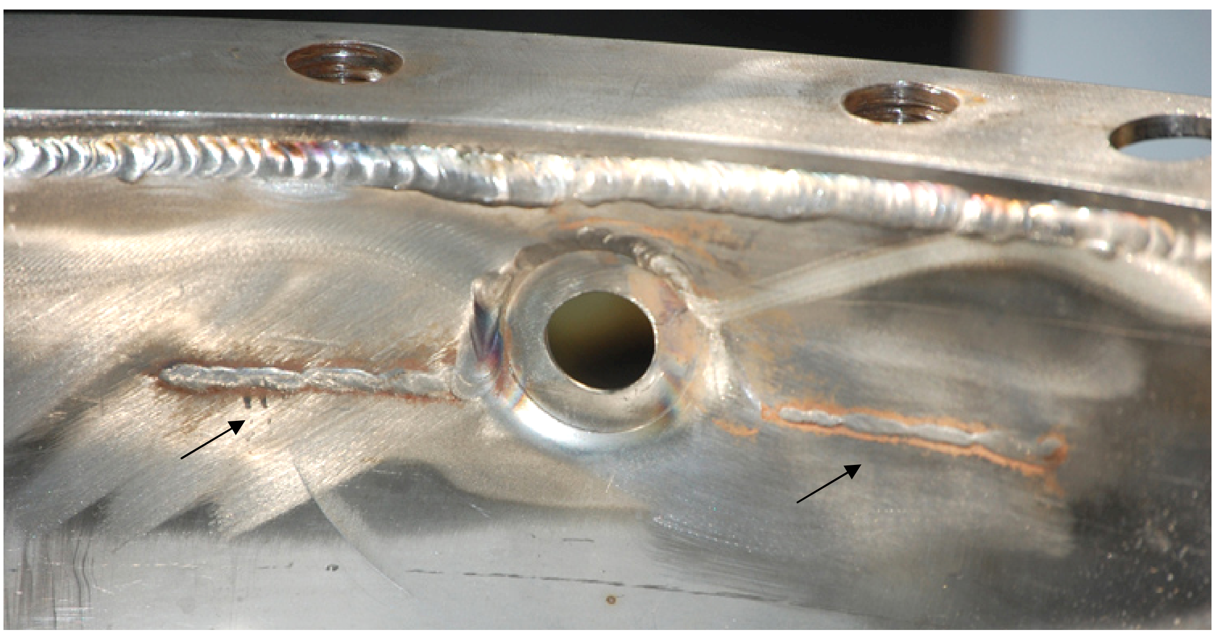


Figure 4. Appearance of light corrosion around the drum flange stitch welds noted after 18 weeks at temperature in phase 2.

(a) 0 degrees (b) 180 degrees (c) 0 degrees (d) 180 degrees

Figure 5. LE1 shield before test (a, b), and after 45 weeks conditioning (c, d).

(a) 0 degrees (b) 90 degrees (c) 180 degrees (d) 270 degrees

Figure 6. LE1 shield after 92 weeks conditioning.

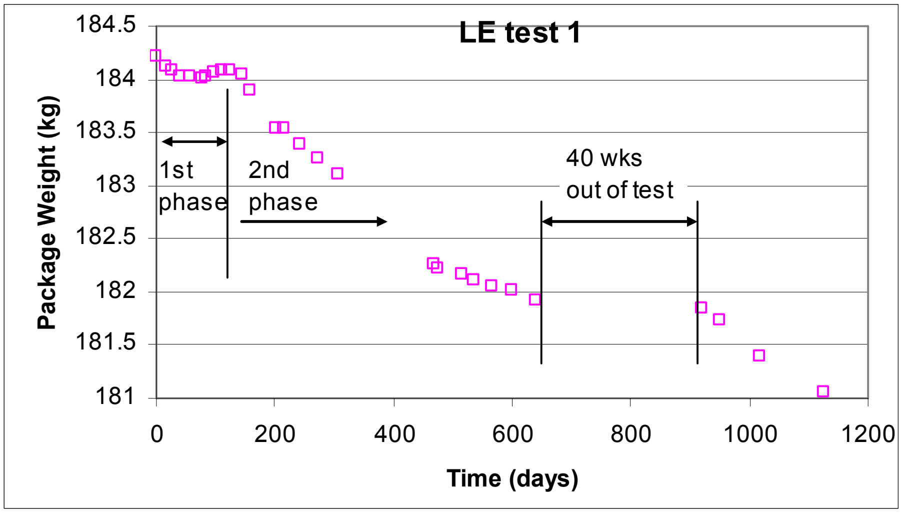


Figure 7. Package weight of LE1 over time.

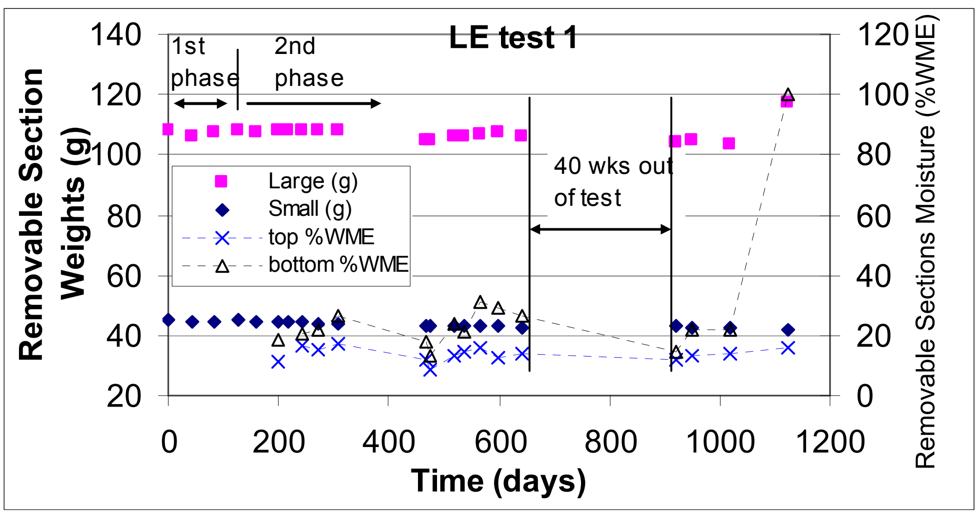
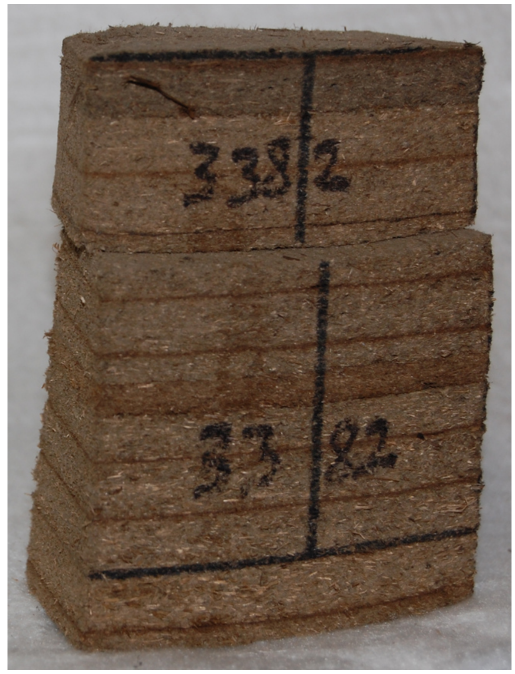


Figure 8. Weight and moisture content of removable fiberboard sections from life extension test 1.

(a) baseline

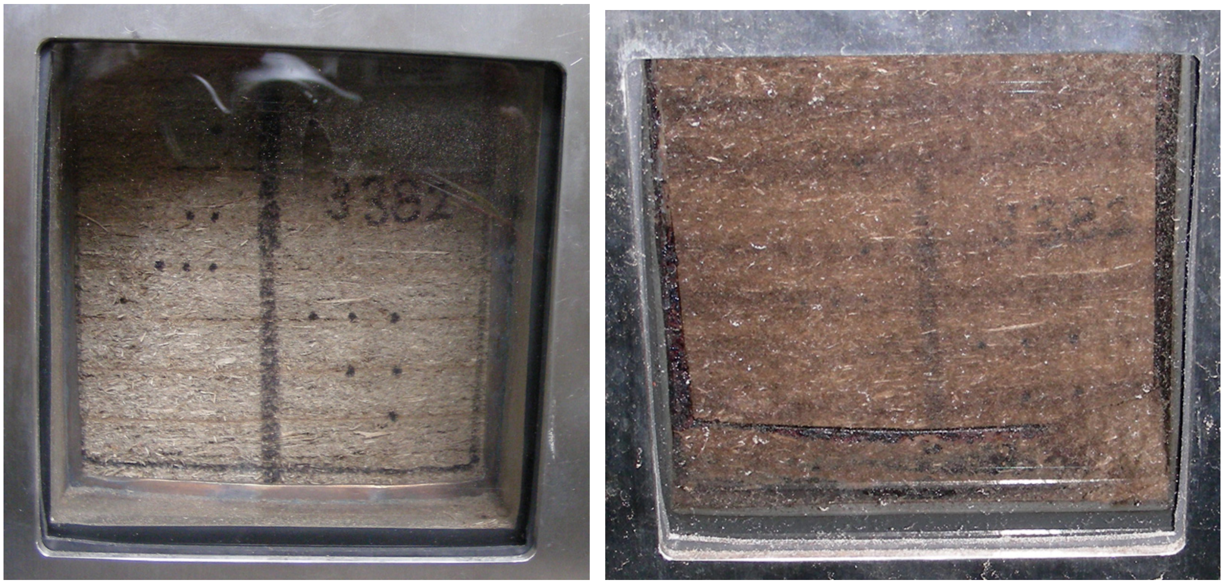


(b) 45 wk conditioning

(d) 63 wk conditioning

(e) 92 wk conditioning

Figure 9. Sequence of photographs of small removable fiberboard sections from test LE1.



(a) baseline (b) after 77 weeks conditioning

Figure 10. View of the side of test LE1 through the view port

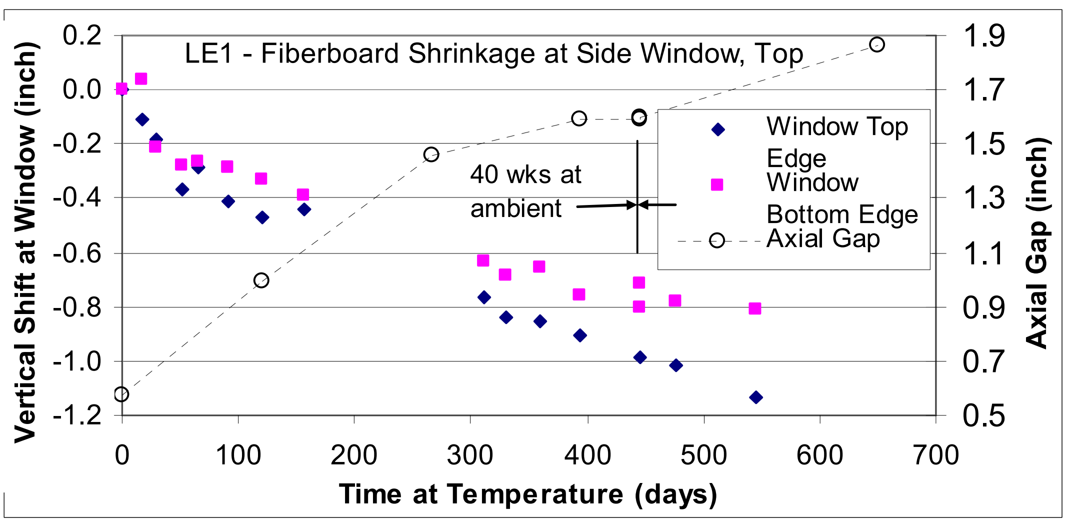


Figure 11. Vertical shift (downward) of fiberboard in LE1, as viewed through the side view port, compared with increase in the axial gap at the top of the package.

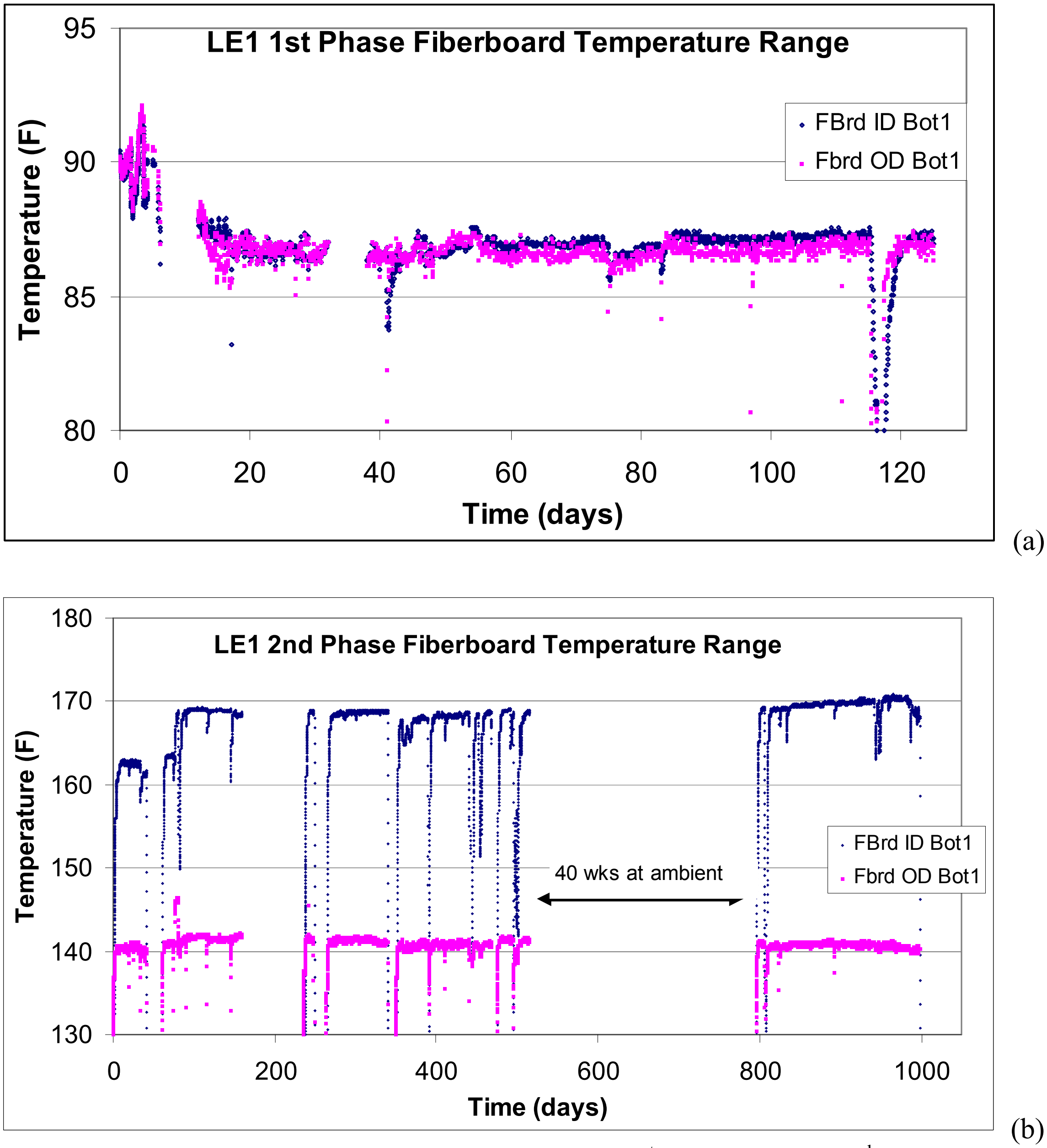


Figure 12. Temperature variation for package LE1 1st phase (a) and 2nd phase (b) at the hottest (blue symbols) and coolest (pink symbols) instrumented fiberboard locations (highest fiberboard ID location and lowest fiberboard OD location – refer to Figure 1).

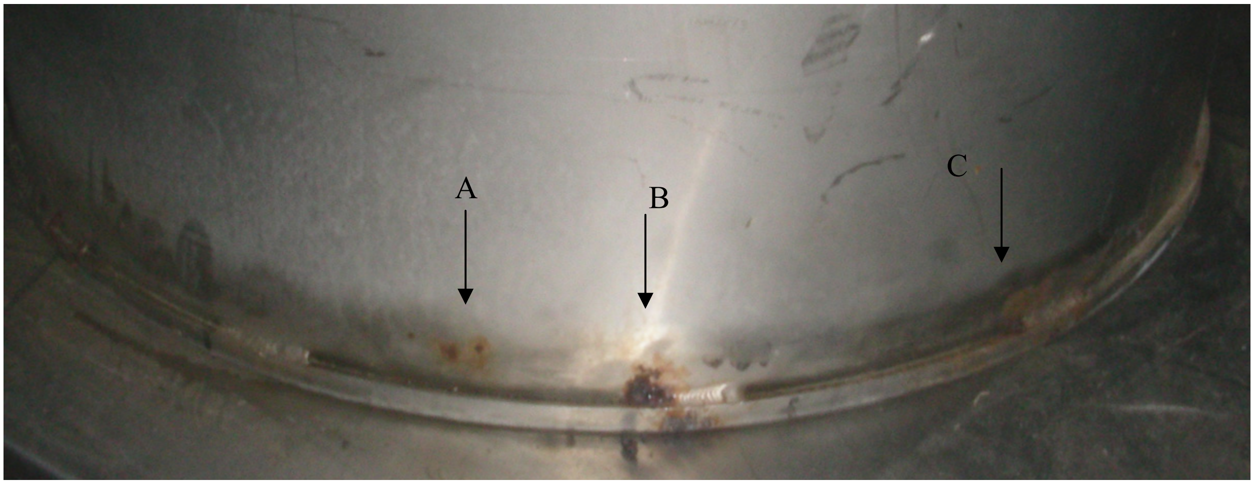


Figure 13. LE1 drum outer surface showing corrosion on (B, C) and near (A) the bottom stitch welds after 92 weeks conditioning.


Figure 14. LE1 drum outer surface at location “A” of Figure 13, before and after cleaning. Permanent marker was added to identify corrosion locations after cleaning.


(a)

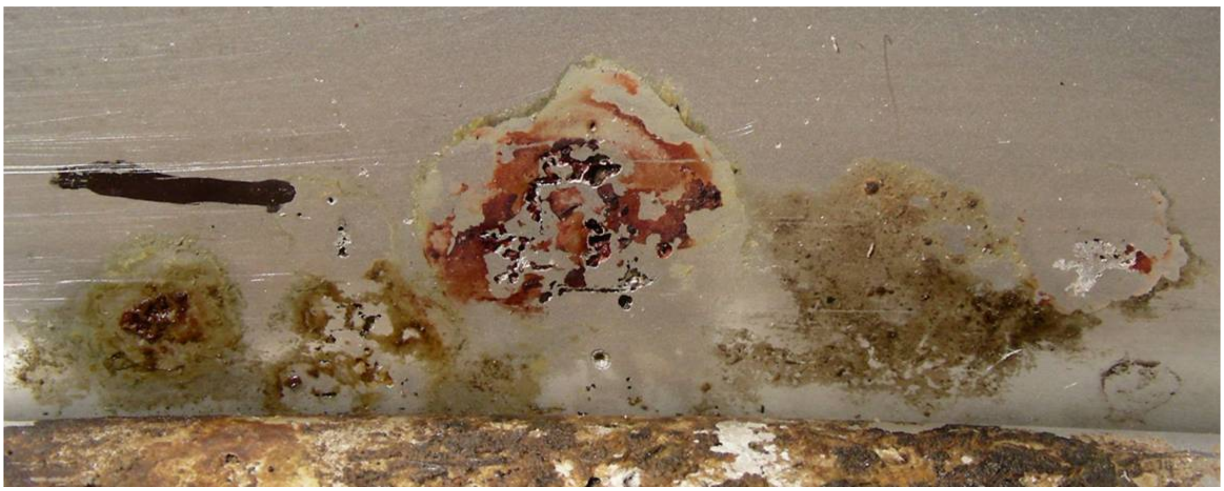

(b)

Figure 15. LE1 drum inner surface behind location "A" (Figure 13), after adherent fiberboard was removed (a), and after further light cleaning (b).

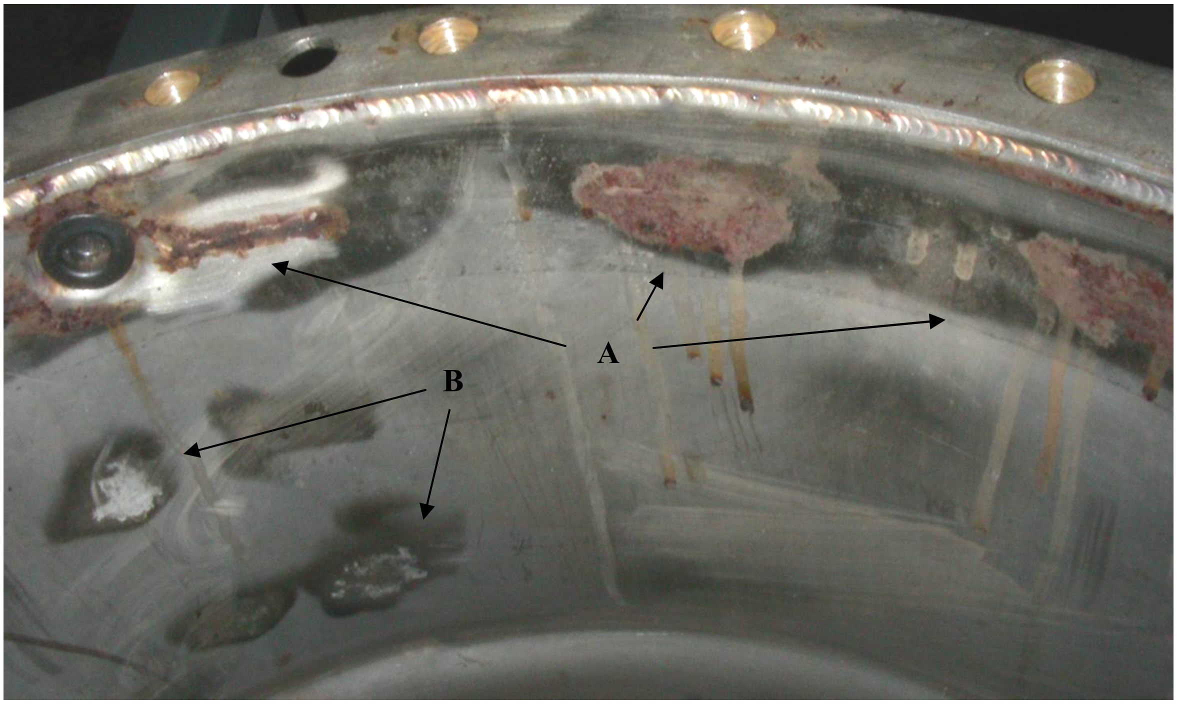


Figure 16. LE1 drum inner surface showing corrosion behind top flange stitch welds (A), and behind bar code plate attachment welds (B).

Figure 17. LE1 drum inner surface behind top flange stitch weld after cleaning.

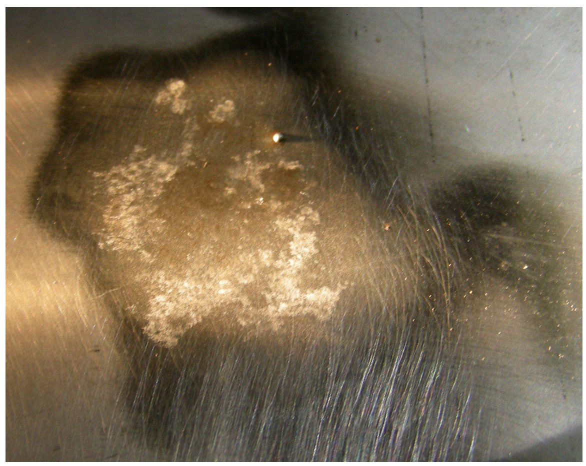


Figure 18. LE1 drum inner surface behind bar code plate attachment weld after cleaning.

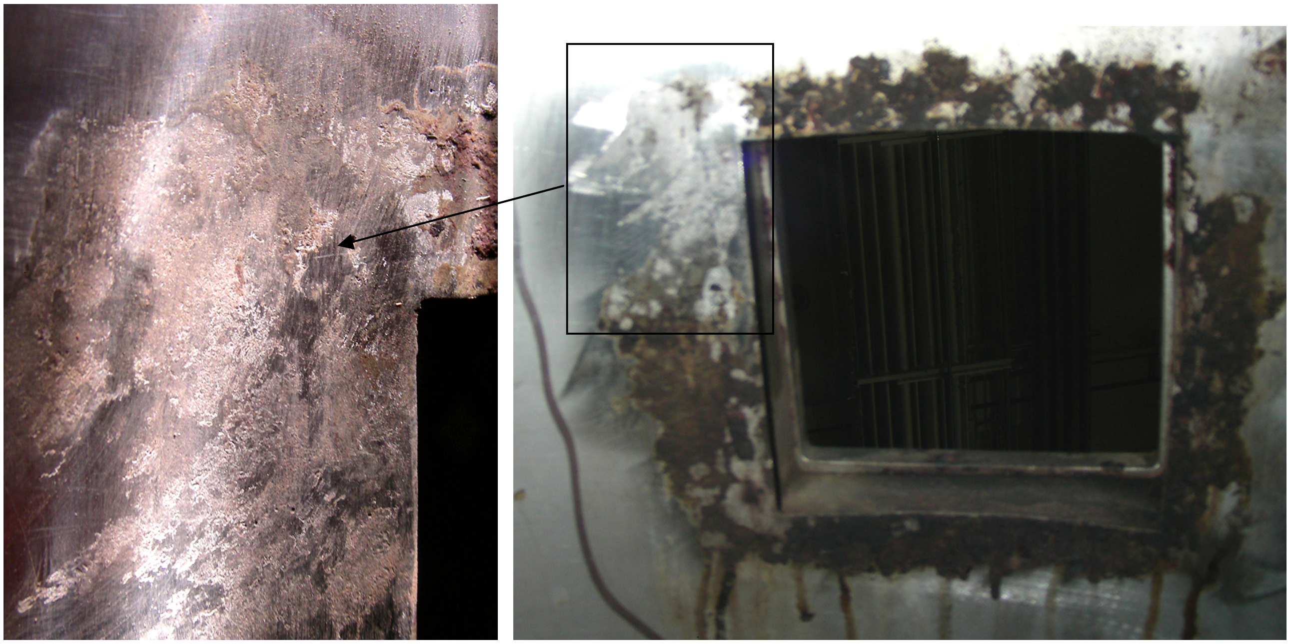


Figure 19. LE1 drum inner surface around window. Corrosion product has been cleaned from one corner revealing general corrosion attack of the drum.

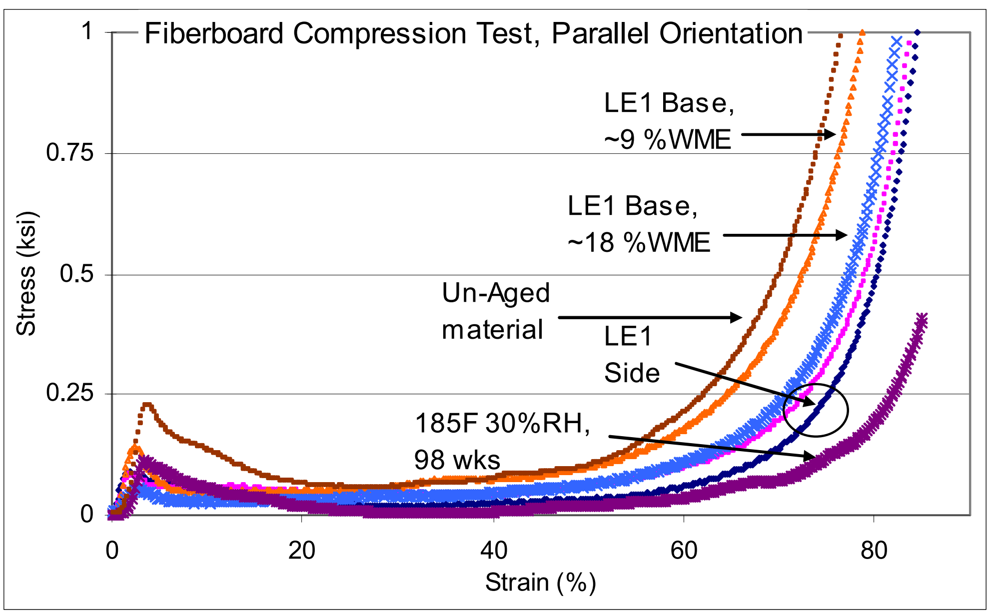


Figure 20. Compression stress-strain curves comparing LE1 fiberboard samples with unaged material and a typical laboratory sample aged a similar time at 185 °F 30%RH, parallel orientation.

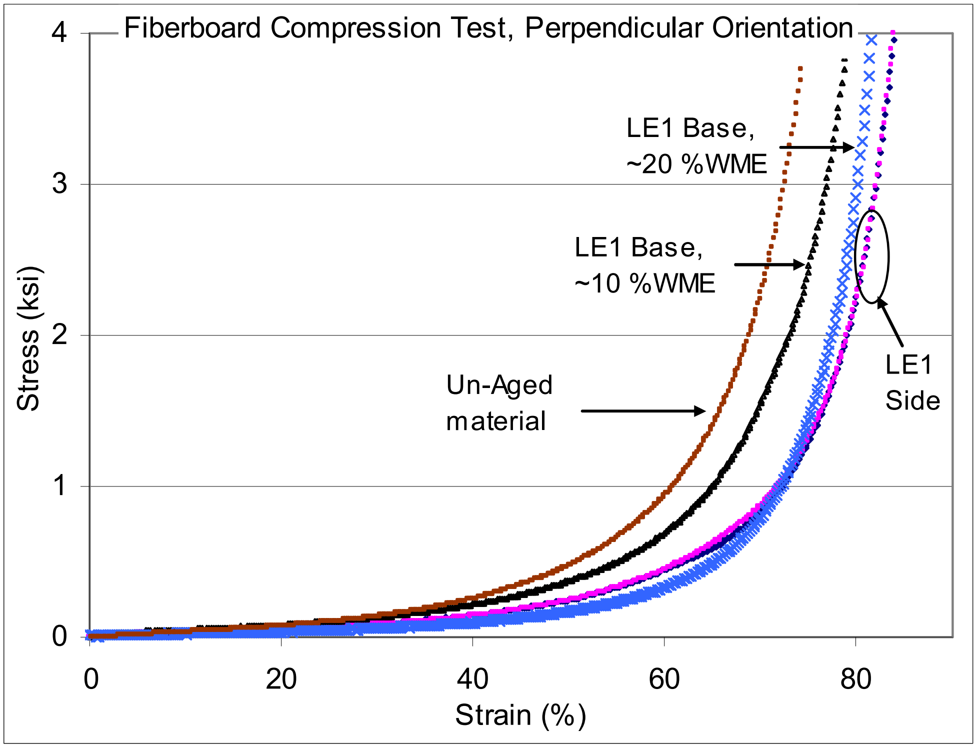


Figure 21. Compression stress-strain curves comparing LE1 fiberboard samples with unaged material, perpendicular orientation.

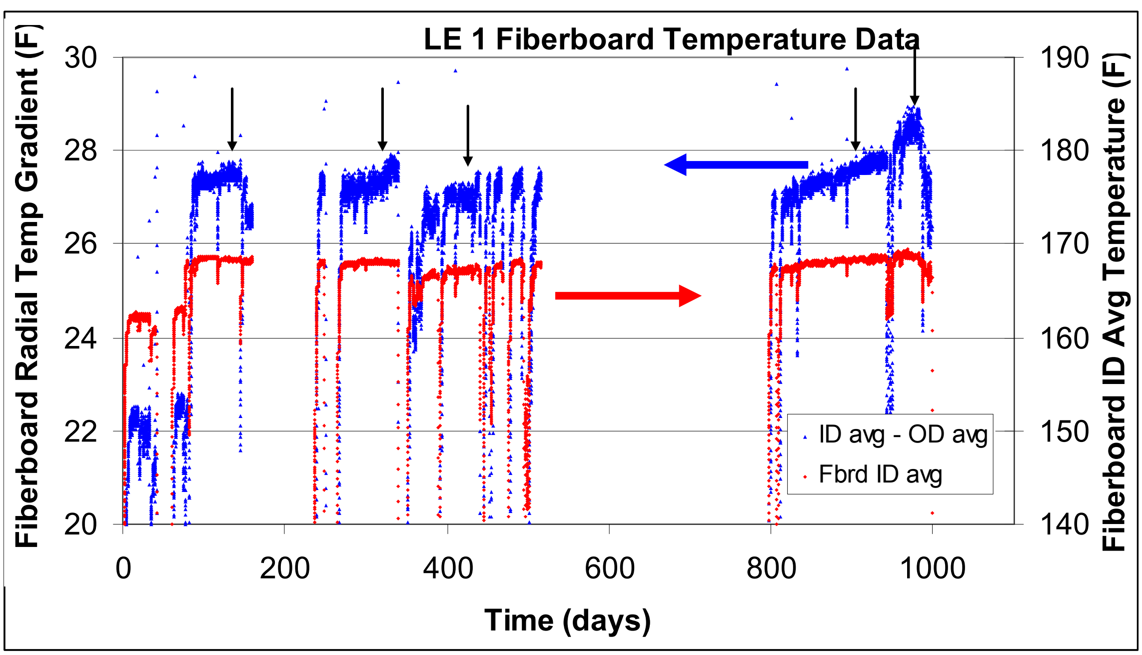


Figure 22. LE package 1 (second phase) average radial temperature gradient (blue symbols) in the fiberboard based on the lower 2 fiberboard ID thermocouples and the lower 2 fiberboard OD thermocouples, and average fiberboard ID temperature (red symbols). The vertical arrows indicate times during steady state operation for which the relative thermal conductivity of the fiberboard was estimated.

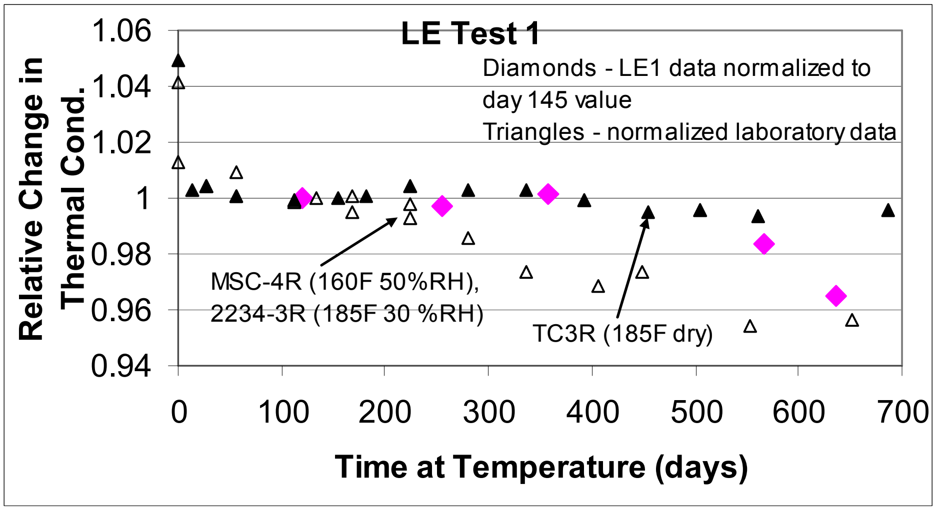


Figure 23. Relative change in radial thermal conductivity estimated from LE1 thermal gradient data (pink diamonds). Comparable normalized data from lab samples following conditioning in 160 °F 50%RH, 185 °F 30%RH and 185 °F dry environments are also shown. The mean test temperature for the laboratory samples was 122 ºF.

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