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9975 SHIPPING PACKAGE LIFE EXTENSION SURVEILLANCE PROGRAM RESULTS SUMMARY

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ABSTRACT

Results from the 9975 shipping package Storage and Surveillance Program at the Savannah River Site (SRS) are summarized for justification to extend the life of the 9975 packages currently stored in the K-Area Complex (KAC). This justification is established with the stipulation that surveillance activities will continue throughout the extended time to ensure the continued integrity of the 9975 materials of construction and to further understand the currently identified degradation mechanisms. The 10 year storage life justification was developed prior to storage. A subsequent report was later used to validate the qualification of the 9975 shipping packages for 10 years in storage. However the qualification for the storage period was provided by the monitoring requirements of the 9975 Storage and Surveillance Program. This report summarizes efforts to determine a new safe storage limit for the 9975 shipping package based on the surveillance data collected since 2005 when the 9975 Storage and Surveillance Program began. The Program has demonstrated that the 9975 package has a robust design that can perform under a variety of conditions. The primary emphasis of the on-going 9975 Storage and Surveillance Program is an aging study of the 9975 Viton® containment vessel O-rings and the Celotex® fiberboard thermal insulation at bounding conditions of radiation, elevated temperatures and/or elevated humidity.

INTRODUCTION

The SRS is storing plutonium (Pu) materials in the K-Area Complex (KAC) [1]. The Pu materials are packaged per the DOE 3013 Standard [2] and shipped to SRS in Model 9975 shipping packages where they are put into interim storage. The 9975 is a robust drum style Type B RAM Packaging meeting all regulatory requirements with a large margin of safety for transportation. Two different models of the 9975 shipping package are used to store plutonium at KAC. The first model is referred to as the -85 package and the second model is called the -96 package. Figure 1 shows a three-dimensional sectional view that is valid for both the -85 and the -96 packages [3,4]. Both shipping packages use an outer 35 gallon drum that is made of 18-gauge Type 304L stainless steel (SS). The external drum has four ½-inch diameter vent holes drilled into its outer perimeter. These holes are located 90 degrees apart, one inch below the drum flange, and each one is fitted with a plastic plug (i.e., a Caplug®). The plugs will burn or melt when exposed to the heat of fire allowing hot gases to escape.

The radiation shielding body for each shipping package is a lead cylinder that surrounds both the primary containment vessel (PCV) and the Secondary Containment Vessel (SCV). Together, the nested PCV and SCV form the double containment assembly. The PCV is a stainless steel pressure vessel that meets Section III, Subsection NB of the ASME code, without the code stamp. The closure assembly is a slightly tapered SS plug that uses redundant O-rings to form a leak-tight seal on the PCV (Figure 2). The SCV is a stainless steel pressure vessel, identical to that of the PCV except for its larger volume, which is designed to accommodate a fully loaded PCV with the closure assembly installed. The O-rings used in both closure assemblies are made from fluorocarbon rubber, specified which is available as Parker Viton® Compound V0835-75 (hereafter referred to as Viton® GLT) or VM835-75 (hereafter referred to as Viton® GLT-S). These two compounds were deemed equivalent based on review of properties and baseline characterization testing.

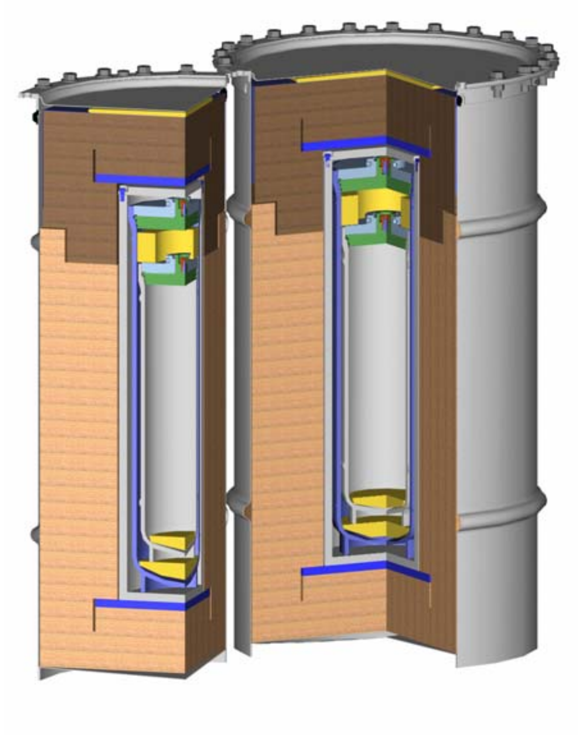
The primary focus of the SRS 9975 Pu Storage and Surveillance program includes the containment vessel O-rings and the insulating material, Celotex® fiberboard. The Celotex® fiberboard is contained between the outer 304L stainless steel drum and the lead shielding in the 9975 package. The fiberboard provides three safety functions: thermal insulation to limit PCV/SCV temperature during a fire [5], criticality control [6] and resistance to package crushing [7] to ensure the plutonium materials remain in a safe configuration during normal and accident conditions [8]. As part of the initial effort to qualify 9975 containers for storage in KAC, the GLT O-rings and Celotex® cane fiberboard materials were assessed to support a 10-year interim storage period [9,10]. The assessment involved a detailed literature review and limited testing of artificially-aged O-rings, along with recommendations for a surveillance program. Subsequently, as a result of material changes, the GLT-S O-ring compound and the Celotex® softwood fiberboard baseline properties were evaluated and found to be comparable with those of the original GLT O-ring and cane fiberboard materials [11]. The primary focus of the SRS 9975 Pu Storage and Surveillance program includes the containment vessel O-rings and the insulating material, Celotex® fiberboard under long-term storage conditions, including bounding conditions of radiation, elevated temperatures and/or elevated humidity.

O-Ring Performance - Containment

Field Surveillance – Actual Conditions:

The predominant degradation mechanism for the containment vessel O-rings in 9975 packages is expected to be compression stress-relaxation or loss of sealing force due to long term exposure at elevated temperatures. Degradation due to radiation is much less likely at the bounding dose rates involved (2 rad/hr or 0.02 Gy/hr). O-rings obtained from field surveillance activities on 195 packages, following up to 8 years of storage in KAC, have shown on average a small degree of compression set (~23% per ASTM D395 Method B) based on nominal dimensions with no other evidence of degradation. The examined O-rings are observed to recover practically all roundness, with hardness and tensile properties similar to those of non-aged O-rings. The O-rings retain high flexibility and resiliency. Visually, no difference has yet been observed between the O-rings removed during field surveillance (~23% compression set) and new O-rings.

For a typical ambient temperature of 85°F, the peak PCV O-ring temperature is ~ 145°F for a maximum 19W payload. For packages with lesser payloads, seal temperatures are reduced. At bounding KAC conditions, peak O-ring temperatures are calculated to reach 200°F. However, these conditions have not been experienced, and would not be sustained on a continuous basis, as the ambient facility temperature changes seasonally. The more moderate temperatures actually experienced to date in KAC ensure that the O–ring integrity remains conservatively bounded by laboratory test data. To date, the 9975 O-ring seals have not been significantly challenged by normal storage conditions in the KAC storage facility.



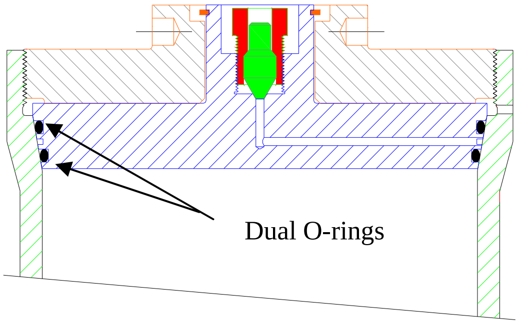


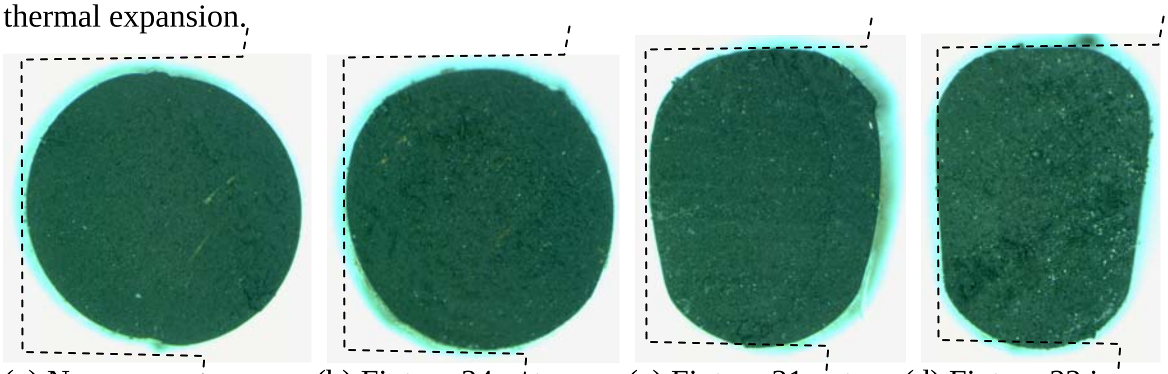
Figure 1. Cutaway of 9975 shipping package Figure 2. Containment vessel seal configuration.

Laboratory Surveillance – Bounding Conditions:

Approximately 63 leak test fixtures were fabricated to the PCV design (mock-up PCVs) and have been periodically helium leak tested per ANSI N14.5 to evaluate leak tightness as a function of aging at elevated temperature. In these tests, both outer and inner O-rings were installed as in real packages, with modifications made to the fixture and test procedure to allow leak testing of both O-rings.

The mock-up PCV fixtures have been aging for up to ~5 years depending on the fixture, its aging temperatures (ranging from 200 to 450°F), and the O-ring type. To date, all of the mock-up PCV fixtures aging at 200°F remain leaktight per the ANSI N14.5 room temperature helium leak test. All fixtures were also tested for permeation to demonstrate that helium has access to the O-rings and to eliminate concern over false positive (leaktight) results [12].

O-rings removed from a fixture after aging at 200°F for 9 months showed a nominal compression set of 18 and 30% 30 days after removal. O-rings aged at 300°F for the same period showed higher compression set (38 – 71% 30 days after removal). These fixtures were still leak-tight at room temperature, but were removed from test for other reasons. In comparison, two fixtures aging at 300°F failed the room temperature leak test after 42 and 44 months at temperature, respectively. The O-rings from these fixtures had compression set of 65 – 89%. Each of these conditions is illustrated in Figure 3. High compression set alone does not mean that an O-ring cannot function or remain leaktight, which is the ultimate criterion for 9975 O-ring performance. The material is still intact, serves as a product release barrier, and may even bond to the mating surface over time, thus maintaining seal integrity [13]. Leak-tightness may even be enhanced at elevated temperature by thermal expansion.



(a) New
0% CS

(b) Fixture 24 outer
23% CS

(c) Fixture 31 outer
65% CS

(d) Fixture 32 inner
89% CS

Figure 3. Cross-section of select O-rings with varying degrees of compression set. The O-ring groove configuration (dashed line) is shown for comparison. The 23% compression set in (b) is typical of field surveillance observations. The O-rings in (c) and (d) were removed from test after they failed to maintain leak-tightness at room temperature (following aging at 300ºF).

It is clear from compression set measurements and visual examination of the O-rings removed from some of the mock-up PCV fixtures that relaxation occurs more rapidly at 300°F. This behavior is believed to be primarily physical in nature (rearrangement of polymer chains) rather than true chemical degradation, though the net effect (loss of sealing force) may be the same.

Laboratory Surveillance – Accelerated-Aging:

Compression stress-relaxation (CSR), is a standard industry metric for evaluating seal performance. Baseline CSR tests performed on the 9975 O-ring compounds (with 25% compression) indicate the seals can lose approximately 90% of initial sealing force at ~500 hours and practically all sealing force at ~1000 hours at 400 °F [14]. This is consistent with the seal manufacturer's generic "continuous" temperature limit of 400°F, as indicated in the Parker O-Ring Handbook [15]. It is important to note that seal performance at any temperature depends on time as well as the seal design. Most design and qualification documents do not address this aspect. Therefore, seal manufacturers strongly recommend end-user testing to confirm such limits.

Longer term CSR data were obtained at aging temperatures of 175-350 °F, providing the basis for a preliminary life prediction model for the V0835-75 O-rings in KAC. This conservative model is based primarily on Arrhenius accelerated-aging methodology. Using the principle of time-temperature superposition, all of the CSR data from different aging temperatures were used to develop a master curve that can be translated to any service temperature. By time-shifting the master curve to various temperatures, the seal lifetime as a function of temperature was developed. (Figure 4) [16-18]. The minimum sealing force required for leaktight performance has not been established, but other investigators have used similar values for critical components and preliminary experiments have shown that such values are reasonable.

At a bounding normal O-ring temperature of 200°F, the model predicts the seals should remain leaktight for at least 12 years. At a more realistic seal temperature of 175°F (106°F ambient) the model predicts a seal lifetime of approximately 20-25 years. A more typical ambient temperature of 85°F reduces the O-ring temperature to ~145 °F at 19 W, which is comparable to the seal temperature resulting from a more typical payload of 12W at an ambient temperature of 106°F (conservative). At the ~145°F O-ring temperature the model predicts a service life of approximately 45-60 years. Given that peak temperatures are not sustained on a continuous basis, the amount of relaxation expected in actual packages is expected to be even less. The time to actual leakage for GLT O-rings at various temperatures is plotted in Figure 5, with life predictions based on CSR data.

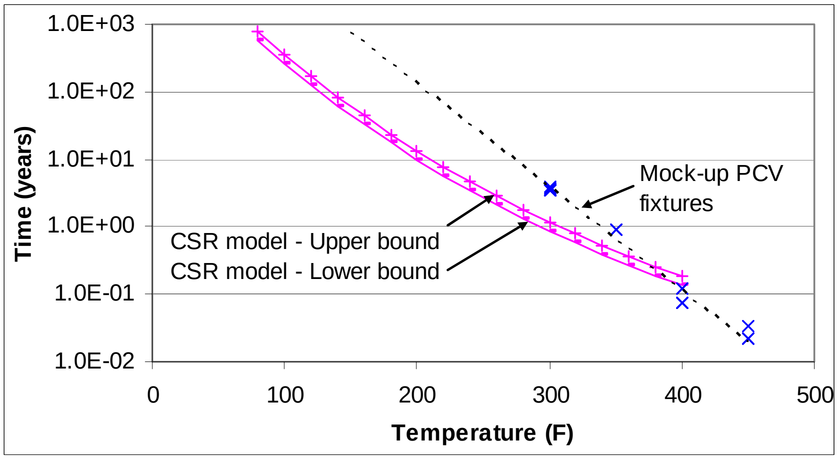


Figure 4. Predicted O-ring lifetime as a function of temperature based on CSR data. The lower bound curve correlates to 90% loss of sealing force. The upper curve prediction correlates to 100% loss of sealing force. Leakage-based service life data for GLT O-rings provided for comparison.

Fiberboard Performance – Fire Resistance, Criticality Control, and Impact Resistance

Field Surveillance – Actual Conditions:

Under normal storage conditions, the anticipated predominant degradation mechanism for fiberboard in 9975 packages is slow pyrolysis due to elevated temperature and/or elevated humidity. This degradation mechanism leads to loss of material, reduced strength, loss of density, decrease in thermal conductivity, reduced assembly height and challenge to criticality controls. The presence of an internal heat source will lead to a moisture gradient within the fiberboard. Regions of elevated moisture can experience enhanced rates of the above degradation mechanisms, as well as the possible growth of mold. Off-normal circumstances which could introduce additional degradation mechanisms include elevated total moisture levels (leading to mold and compaction) and infestation (leading to fiberboard consumption).

After several years of storage in KAC, only one shipping package during the NDE process was found with mold on the fiberboard (Figure 6). Further inspection identified regions of mold on both the upper and lower fiberboard assemblies, elevated moisture levels in the regions of mold, and the axial gap above the fiberboard exceeded the 1 inch maximum criterion (consistent with compaction of the lower fiberboard layers). Twelve additional packages in KAC have been identified through activities other than NDE that failed the axial gap criterion. In a majority of these packages, the fiberboard contained mold and regions of elevated moisture levels. The moisture levels were generally highest in the bottom layers, leading to compaction and the increased axial gap.

Figure 6. Example of moldy fiberboard seen on 9975-01710 from KAC.

Laboratory Surveillance – Bounding Conditions:

Laboratory surveillance has been performed on small-scale fiberboard samples to track changes in the mechanical, thermal and physical properties during short and long-term aging. Aging environments were selected to provide conditions typical of KAC service as well as bounding to that service. Following baseline data, the maximum exposure periods in the various environments range from 17 to 287 weeks (through January 2011).

The highest temperature environment is 250°F, which is the operating limit identified for fiberboard in the 9975 SARP [3,4]. Current calculations show that portions of the fiberboard could approach this value only during extreme conditions, although these events would be relatively short-term. In the more moderate temperature environments, elevated humidity levels are combined with elevated temperature. For the tested environments, short-term exposures tend to produce property changes based primarily on changes in sample moisture content. The fiberboard readily gains or loses moisture in response to the surrounding environment, and the moisture content has a strong influence on fiberboard properties. The longer-term data show the impact of material degradation.

While the moisture content of fiberboard will tend to seek equilibrium with the humidity of the surrounding air, the 9975 drum provides a significant degree of isolation between the fiberboard and the external environment. An upper assembly exposed directly to the ambient air experiences a variation in weight of ~1.5% as a result of seasonal humidity changes. When placed inside a closed 9975 drum, however, the variation is reduced approximately 50X. The moisture which is contained in the fiberboard when it is placed in the drum will likely define the humidity level inside the drum for a long time.

Mechanical Properties

Mechanical strength of the fiberboard is required primarily for energy absorption during an impact or drop event. The compression test produces a stress-strain curve, with the area under the curve being proportional to the energy absorbed. The area under the stress-strain curve up to a strain of 40% is used as a metric for comparing results between samples. Figure 7 shows the variation in this metric over time for samples conditioned at several elevated temperatures, and for samples conditioned at 185°F (85°C) with several humidity levels. Noticeable declines over time in energy absorption capability are seen for elevated temperatures of 215°F and greater, and for elevated humidity levels at 185°F. The Figure 7 data apply to material tested in the parallel orientation (typical of a package crushed from the side). The same trends are observed, although at a slower rate, for material tested in the perpendicular orientation (typical of an impact to the top of a package).

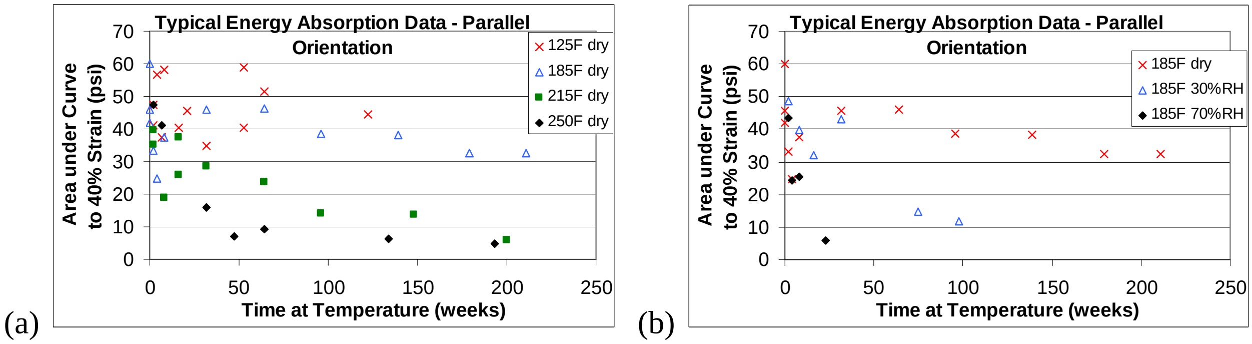


Figure 7. Area under the compression stress-strain curve up to 40% strain, for parallel orientation samples. All samples shown are from a single package source designation to eliminate package-to-package variation.

Thermal Properties

Thermal conductivity of fiberboard samples is measured at three mean temperatures – 77, 122 and 185°F (25, 50 and 85°C). The same trends are seen in thermal conductivity for each test temperature. Thermal conductivity is also measured for two orientations – with heat flow parallel to the fiberboard layers (radial) and with heat flow perpendicular to the fiberboard layers (axial). Significant changes in thermal conductivity result from conditioning in elevated temperature and humidity environments, as shown in Figure 8. While the elevated humidity exposures are of relatively short duration, the rate of degradation from each environment is seen in the slope of the data for each data set.

In Figure 8 (a), the step decrease between thermal conductivity at time 0 and subsequent data reflects a change in the moisture content of the samples. The initial exposure to elevated temperature drives off most of the moisture within the material, with a resulting decrease in the thermal conductivity. This change is reversible if the material is allowed to re-absorb moisture. However, the decrease in thermal conductivity during extended exposure to the aging environments is not reversible.

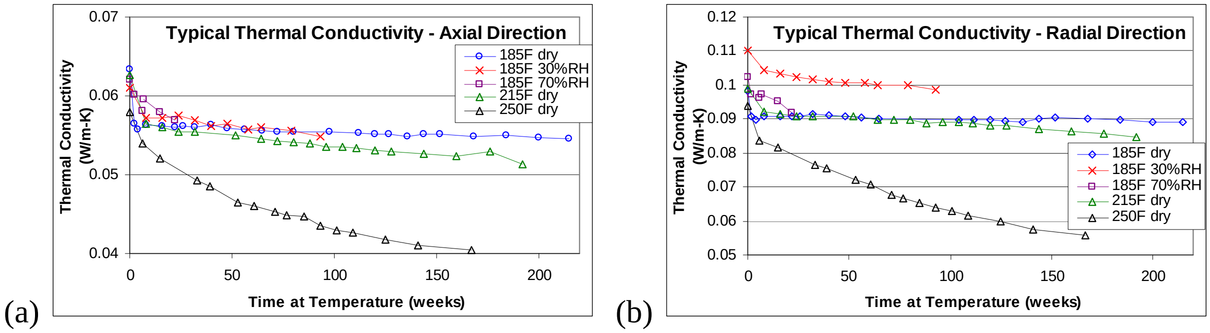


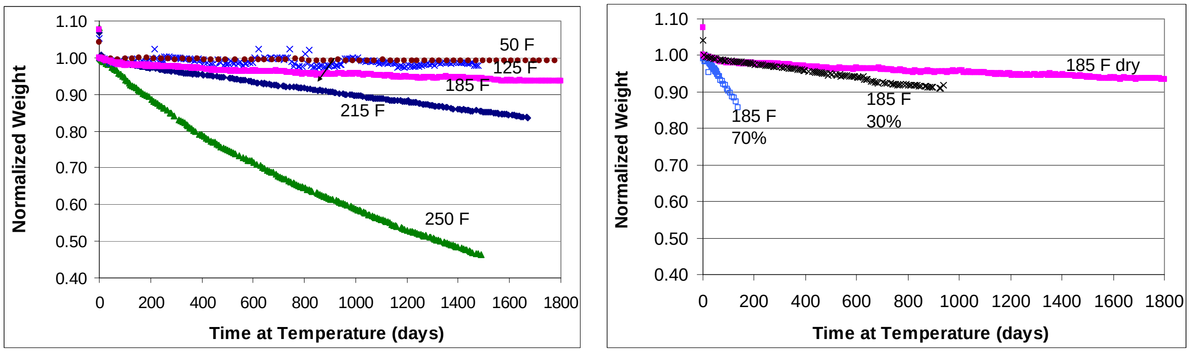
Figure 8. Thermal conductivity data for 77F mean temperature following conditioning in several environments. (a) axial orientation, (b) radial orientation.

The specific heat capacity is measured following different intervals in various environments, including elevated temperature and/or humidity. In general, the specific heat capacity data show significant scatter, but do not show significant degradation [19].

Physical Properties

Fiberboard weight, dimensions and density change as a result of change in moisture content and/or aging. These properties for samples aged in several environments are summarized in [19]. For example, Figure 9 shows the weight change experienced in several dry and humid environments. In this figure, the weight is normalized to its initial conditioned value to provide a common point of comparison across different samples.

Among the nominally dry environments, each of the physical properties degrades faster as the temperature increases. At a given temperature, each property degrades faster at higher humidity levels. However, there may be a humidity threshold below which the humidity level does not affect the degradation rate. The difference in physical properties is small for samples conditioned in a 185°F dry oven and a 185°F 30% humidity chamber.



(a) (b)

Figure 9. Relative weight change for fiberboard samples conditioned in several environments. Samples in (a) were conditioned in dry environments, at the temperatures noted. Samples in (b) were conditioned at 185°F, at the humidity levels noted.

Other

Limited testing to date on full 9975 packages shows that some environments can have a strong influence on package performance. In particular, as the internal heat load creates a thermal gradient within the fiberboard, the moisture in the fiberboard will also form a gradient. In general, higher moisture levels will develop in the cooler regions of the fiberboard. In extreme conditions, if regions of the fiberboard are heated high enough, the water in those regions will evaporate, but the water vapor will not easily escape the drum. Therefore, it will tend to condense in the coolest portions of the drum. The temperature distribution will define the extent of moisture redistribution. This behavior has been observed in three instrumented test packages that were heated internally as well as from the outside. Each package started with typical fiberboard moisture levels. In all three cases, the bottom fiberboard layers eventually became saturated with moisture.

CONCLUSIONS & OBSERVATIONS

Surveillance data have been obtained on 780 field O-rings taken from 195 Model 9975 packages after various storage periods and show no signs of O-ring degradation. Mock-up PCV fixtures with O-rings aged at 200°F for 5+ years on a continuous basis remain leaktight. The leak testing approach provides an excellent method for monitoring O-ring performance real-time at bounding conditions. These results indicate longer-term stability at lower temperatures. Accelerated-aging tests using CSR as a parameter to measure seal performance indicate that the O-rings are capable of retaining measureable sealing force for many years if not decades at realistic storage conditions. As non-Arrhenius aging behavior may be possible due to long-term oxidation or other mechanisms, additional testing is in progress to determine if a change in the degradation mechanism will occur.

Degradation of physical, mechanical and thermal fiberboard properties in compliant packages at storage temperatures is generally slow and will provide significant time before required margins are compromised. Fiberboard properties measured on packages following up to 7 years in storage indicate the material to have experienced no significant degradation. It is further expected that these packages would continue to show no significant degradation for at least 15 years in storage. Accordingly, based on laboratory testing, compliant packages stored under such conditions are expected to continue to meet minimum requirements for KAC for thermal resistance, criticality control, and impact resistance for up to 15 years in storage.

Based on the data obtained to date and projected performance of the 9975 packages in the KAC facility, the 9975 packages are expected to maintain the minimum safety requirements for a period of at least 15 years in KAC storage. This justification is established with the stipulation that surveillance activities will continue throughout this extended time to ensure the continued integrity of the 9975 materials of construction and to further understand the currently identified degradation mechanisms.

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