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Properties of Fiberboard Overpack Material in the 9975 Shipping Package Following Thermal Aging

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Abstract
Many radioactive material shipping packages incorporate cane fiberboard overpacks for thermal insulation and impact resistance. Mechanical, thermal and physical properties have been measured on cane fiberboard following thermal aging in several temperature/humidity environments. Several of the measured properties change significantly over time in the more severe environments, while other properties are relatively constant. These properties continue to be tracked, with the goal of developing a model for predicting a service life under long-term storage conditions.

INTRODUCTION
The Savannah River Site (SRS) uses 9975 shipping packages to store plutonium and uranium materials in the K-Area Materials Storage (KAMS) facility [1]. Although the 9975 is a certified shipping package, it continues to perform several safety functions in KAMS, including criticality prevention, impact resistance, content containment, and fire resistance. The 9975 is used as a component of KAMS to ensure the stored materials remain in a safe configuration during facility normal and accident conditions. Celotex® brand cane fiberboard, manufactured by Knight-Celotex, is contained between the outer stainless steel drum and the inner lead shielding in the 9975 package. The fiberboard used in 9975 packages meets ASTM C208-95, Grade IV wall sheathing. Layers of fiberboard are laminated together with water-based polyvinyl acetate (PVAC) adhesive.

The 9975 package is expected to perform its safety functions for at least 12 years from initial packaging (2 years in transport + 10 years in storage). Uncertainty exists concerning the stability of the cane fiberboard over time in the KAMS storage environment. Testing is being performed to predict a service life for this material, and to validate satisfactory physical, mechanical and thermal performance over that service life. This paper describes findings from testing of cane fiberboard after conditioning at specific temperature and humidity levels.

The maximum normal operating temperature in the KAMS storage area is 120°F. Steady state thermal analysis of the 9975 package predicts this would produce a maximum internal fiberboard temperature of 184°F. Off-normal or accident conditions can produce higher fiberboard temperatures, up to 250°F. The 9975 shipping package safety analysis allows fiberboard operating temperatures of up to 250°F. Since air / moisture can infiltrate the 9975 drum, the insulation can be exposed to moisture levels in equilibrium with the ambient atmosphere. This would produce relative humidity levels up to 70% (at 133°F) or 20% (at 184°F), based on published psychrometric charts.

Test Plan

The fiberboard material must retain its dimensions and density within certain ranges to provide the required impact resistance, criticality spacing and fire resistance. Several properties of interest to demonstrate acceptable long-term performance of the material include dimensional stability, moisture absorption/retention, density, compressive strength, thermal conductivity and specific heat capacity.

Baseline testing of mechanical and thermal properties has been performed at temperatures of 25, 51 and 91ºC (77, 125 and 195ºF) [2, 3]. Subsequent testing has been performed following environmental exposures of up to 72 weeks. Conditioning environments include nominally dry environments at 51 to 121ºC, and humidity controlled (50 or 70% relative humidity) environments at 51 to 85ºC.

Test samples were taken from several different packages, and includes the following:
- undamaged portions of fiberboard assemblies from drop tested packages (LD1, LD2, MSC), which have been in storage for ~10 years
- lower assembly from an unused package following several years storage (KT2)
- lower assemblies removed from packages following several years service (2234, 826)
- remnant portions of a new assembly purchased for a separate effort (NEW)

Conditioning and testing will continue into 2008, to provide a maximum body of data prior to establishing a model for predicting a service life in KAMS. Preliminary analysis of data provides confidence that meaningful predictive models can be generated with the additional data. Analysis also led to identifying additional samples that could be added to the conditioning environments to fill in weak portions of the original test matrix.

Compression Test Samples

Compression samples, approximately 5 x 5 x 5 cm, were tested with the applied load either parallel or perpendicular to the fiberboard layers following environmental exposure. Testing was performed at the conditioning temperature, but without humidity control.

The mechanical behavior of the fiberboard can be characterized in a number of ways. Of greatest importance to the performance of the 9975 package is the energy absorption capacity. Two metrics used to characterize mechanical performance are the area under the stress-strain curve up to a strain of 40% (applicable to all samples) and the buckling strength (applicable to parallel orientation samples).

Thermal Test Samples

Thermal conductivity samples are typically about 30 x 30 x 3 cm, although some samples are ~18 x 18 x 3 cm. Thermal conductivity is measured on fiberboard samples using a Lasercomp Fox 300 instrument. This instrument provides results consistent with ASTM C518. The thermal conductivity is measured at up to 3 mean temperatures. All samples are measured at mean temperatures of 25 and 51ºC. Samples conditioned at 71ºC are also tested at that temperature as well. Samples conditioned at 85ºC or higher are also tested at a third temperature of 85ºC.

Specific heat capacity samples are approximately 2.5 cm diameter and 5 cm high. This testing is performed in accordance with ASTM C351 at mean test temperatures of 25 and 51ºC. At both mean temperatures, there is significant scatter in the data.

Physical Measurement Samples

Physical measurements (dimensions, weight) are made on samples approximately 5 x 5 x 5 cm. Dimension and weight data have been collected at 51 to 121ºC, either in a nominally dry oven, or in a humidity controlled environment. A limited number of samples began conditioning at low temperature (approx. -10 and 10ºC / 14 and 50ºF) to determine if the dimensions or density change at low temperature. At 10ºC, samples were conditioned dry (with desiccant), at ambient humidity, and at ~100% humidity. At -10ºC, samples were conditioned dry (with desiccant) and at ambient humidity.

As a comparison to these data, the weight and dimensions of compression and thermal conductivity samples are recorded prior to testing. The compression samples are measured only twice (prior to conditioning and prior to testing), while the thermal conductivity samples are measured a number of times since they are tested repeatedly. The limited handling of these samples would provide less opportunity for inadvertent weight loss during handling.

Three fiberboard upper subassemblies are weighed and measured periodically. Following the accumulation of greater than 12 months data, and establishing the seasonal variation in these measurements, one of the upper subassemblies was placed back inside its drum and removed only for monthly measurement. Subsequently, a second upper subassembly was placed back inside its drum and will be removed for measurement after a much longer interval. This is intended to identify the degree of isolation provided by the drum.

Test Data

Compression Data

A significant decrease in strength is seen at 121C, with greater decreases following longer exposure times. At lower exposure temperatures (102C and less) and nominally dry, there is no significant degradation. Some decrease in strength is seen at 85C, 70% RH over 12 weeks. There is no significant degradation in the humidity controlled environments at lower temperatures. These trends are consistent for each material source. Typical compression test stress-strain curves are shown in Figures 1-3.

The degradation in strength at 121C is seen in both the buckling strength for samples tested in the parallel orientation (Figure 4) and in the area under the stress-strain curve to 40% strain (Figure 5).

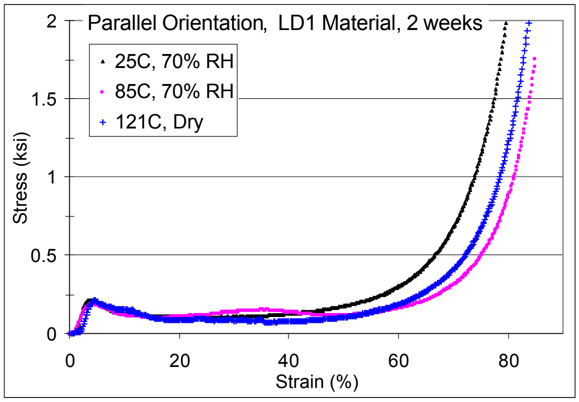


Figure 1. Compression stress-strain curves for typical samples after 2 weeks exposure, parallel orientation.

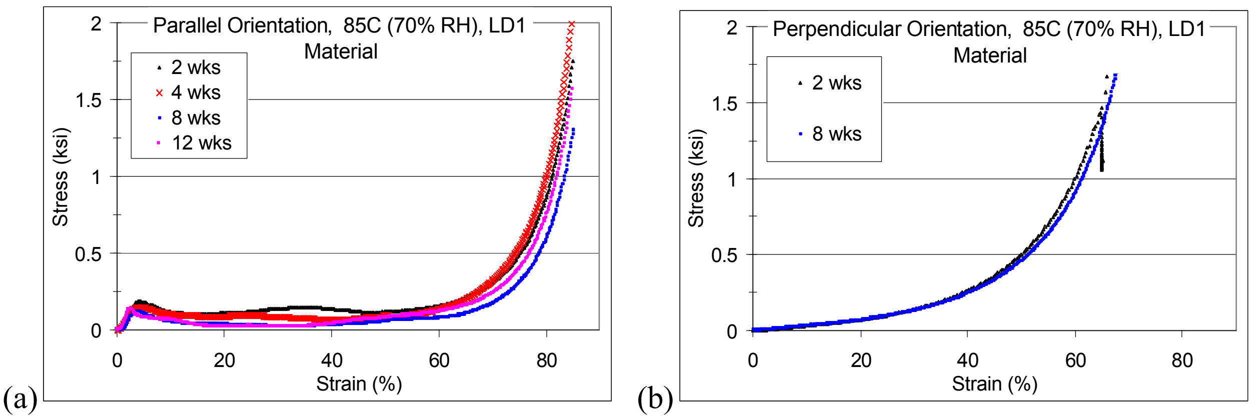


Figure 2. Compression stress-strain curves for LD1 material conditioned at 85C, 70%RH, (a) parallel orientation and (b) perpendicular orientation.

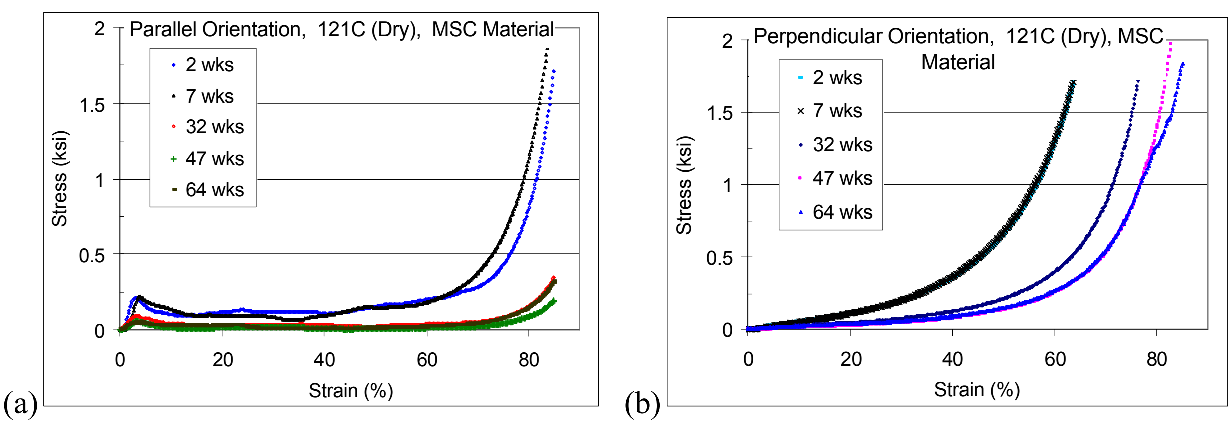


Figure 3. Compression stress-strain curves for MSC material conditioned at 121C, dry, (a) parallel orientation and (b) perpendicular orientation.

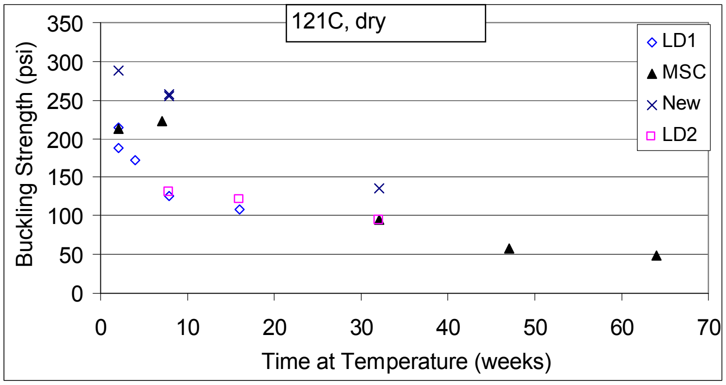
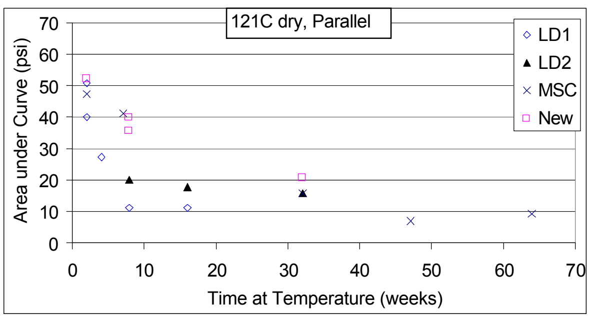


Figure 4. Buckling strength for compression samples tested in the parallel orientation, following conditioning at 121C.

(a) 

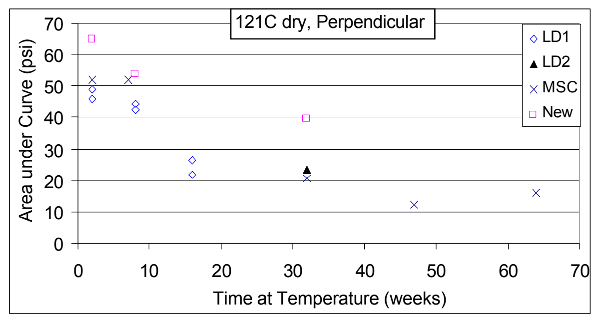
(b) 

Figure 5. Area under stress-strain curve up to 40% strain, following conditioning at 121C. (a) parallel orientation, (b) perpendicular orientation

Thermal Data

Thermal conductivity data are summarized in Figures 6-9. Each environment includes samples that were prepared to allow heat flow during testing in either the radial or axial direction. For the radial direction, heat flow is parallel to the fiberboard layers, representing heat flow radially through the 9975 package. For the axial direction, heat flow is perpendicular to the fiberboard layers, representing heat flow axially through the 9975 package.

An initial drop in thermal conductivity (from 0 to 2 weeks exposure) reflects a loss of moisture in the samples. At 121C, dry, the thermal conductivity continues to decrease with time for both orientations. At 85C, dry, there is no significant change in thermal conductivity following the initial drop, for exposures through 72 weeks. In contrast, Figure 8 shows data for samples that were held at 85C but cycled between a nominally dry oven and 70% RH. The data within the rectangles were taken after exposure to 70% RH, and reveal two distinct trends. First, the thermal conductivity tends to increase in the more humid environment as a result of moisture absorption. The adsorption is also reflected in a weight gain. Second, with continued exposure to 70%RH, the thermal conductivity decreases over time.

Similar to the samples at 85C, a set of samples was also cycled between a nominally dry oven and 70% RH at a constant temperature of 51C. For these samples, a modest increase in thermal conductivity and weight gain is seen following 70% RH exposures. However, there is no significant change in thermal conductivity with continued exposure to the humid environment.

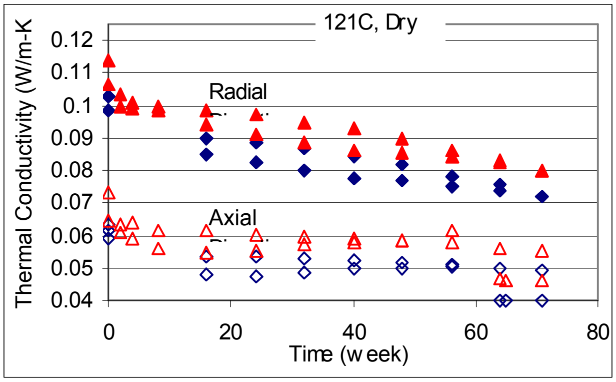


Figure 6. Thermal conductivity of fiberboard following conditioning at 121C, dry, and tested at a mean temperature of 25C (blue symbols) and 85C (red symbols).

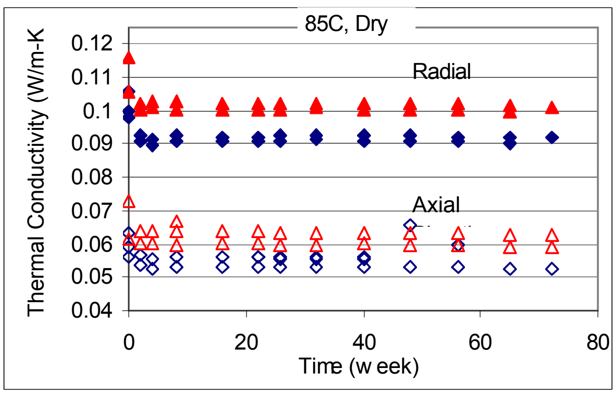


Figure 7. Thermal conductivity of fiberboard following conditioning at 85C, dry, and tested at a mean temperature of 25C (blue symbols) and 85C (red symbols).

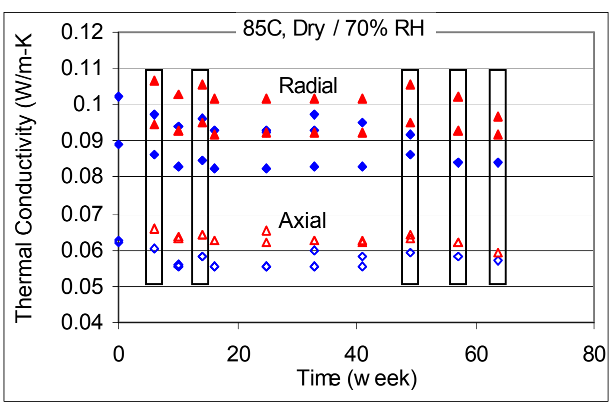


Figure 8. Thermal conductivity of fiberboard following conditioning at 85C, dry / 70%RH and tested at a mean temperature of 25C (blue symbols) and 85C (red symbols). Data within the rectangles follows exposure at 70%RH. The remaining data follows nominal dry exposure.

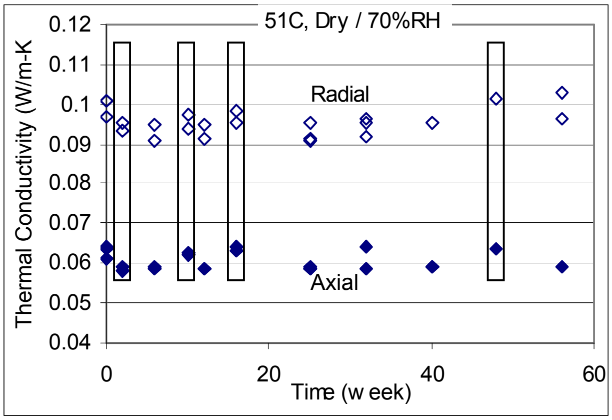


Figure 9. Thermal conductivity of fiberboard following conditioning at 51C, dry / 70%RH and tested at a mean temperature of 25C. Data within the rectangles follows exposure at 70%RH. The remaining data follows nominal dry exposure.

The specific heat capacity was tested on multiple samples following exposures to the same environments as the thermal conductivity samples. These test data show a significant degree of scatter, but no overall trend with time. Average specific heat capacity values through 64 weeks exposure in most environments typically fall between 1200 and 1500 J/kg-K.

Dimensional and Density Data

Dimensional and weight information have been collected on several samples in a number of environments. Since each sample has a different initial weight, the data is compared on the basis of relative weight change. All data is normalized to the sample weight after the first measurement following exposure environment. This eliminates the effect of initial change from moisture loss, and allows multiple samples to be easily compared. Weight loss data for several environments are shown in Figures 10-12.

Weight loss is observed with time in all the elevated temperature environments. No significant change is observed in the cold environments. The greatest rate of weight change is seen at 85C, 70% RH. Among the dry environments, the higher temperatures produce a greater degree of weight loss. Similar trends are observed for each of the material sources, although the longest term data are for the MSC material.

The density changes in a manner similar to the weight. The greatest rate of change occurred at 85C, 70% RH. However, the magnitude of change is not as great since the sample dimensions change along with the weight. For samples at 121C, the weight decreases by about 14% after 400 days, while the density decreases by about 9% in the same period.

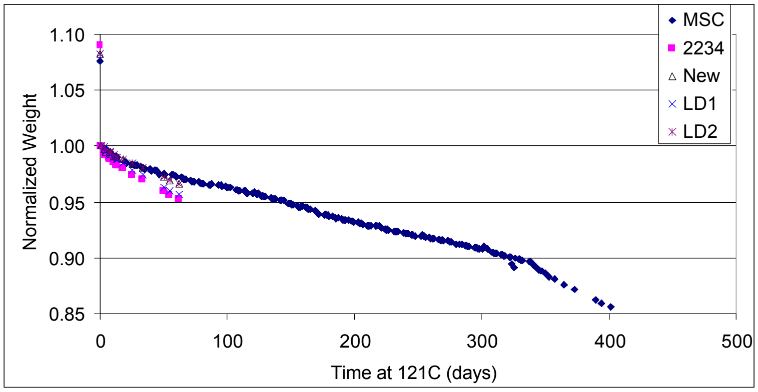


Figure 10. Relative change in weight for samples conditioned at 121C, dry.

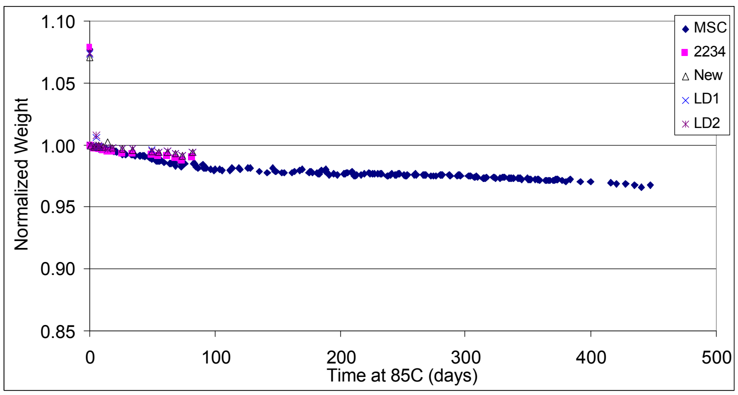


Figure 11. Relative change in weight for samples conditioned at 85C, dry.

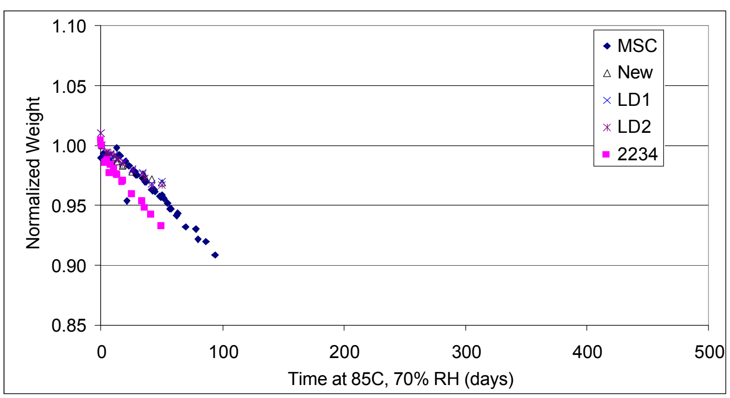


Figure 12. Relative change in weight for samples conditioned at 85C, 70% RH.

The weight and dimensions of two upper fiberboard assemblies have been tracked for over a year. Both assemblies have been in the same (ambient) environment and displayed the same seasonal variations in weight. After the degree of seasonal variation was established, one assembly was returned to its 9975 drum and continued to receive regular measurements. The degree of weight variation decreased significantly. To further quantify the degree of isolation provided by the drum, the second assembly was returned to its 9975 drum and will not be measured until the seasonal variations in a third control assembly reach a maximum or minimum value. Comparing these assemblies should indicate the degree of air/moisture exchange provided by the closed drum versus the degree of air/moisture exchange provided each time the drum was opened for measuring the assembly. The weight variations of the upper assemblies are shown in Figure 13.

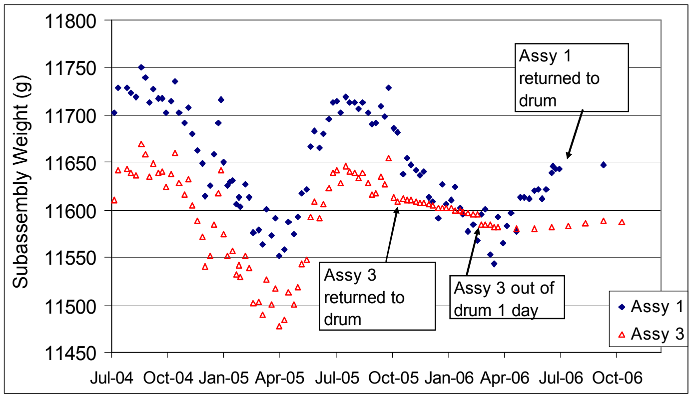


Figure 13. Seasonal weight variations of two upper fiberboard assemblies exposed to ambient conditions, and following return to a 9975 drum.

General Observations

In addition to the specific data mentioned above, there are several general observations to note. The visual appearance of many samples changed during conditioning, particularly at the higher temperatures and/or elevated humidity. The most obvious visual change is a darkening of the sample. Some of the samples at 121ºC also developed very dark spots across their surface. After extended periods at elevated temperature (particularly at 121ºC) and low humidity, samples typically developed a feel of being weak and brittle. For compression samples, this also corresponded to a tendency for the sample to fracture and split apart when tested in the perpendicular orientation, and at longer conditioning periods compression samples tested in both orientations tended to crumble to a powder rather than maintain the integrity of individual fibers. Some of these conditions are shown in Figures 14 - 15.

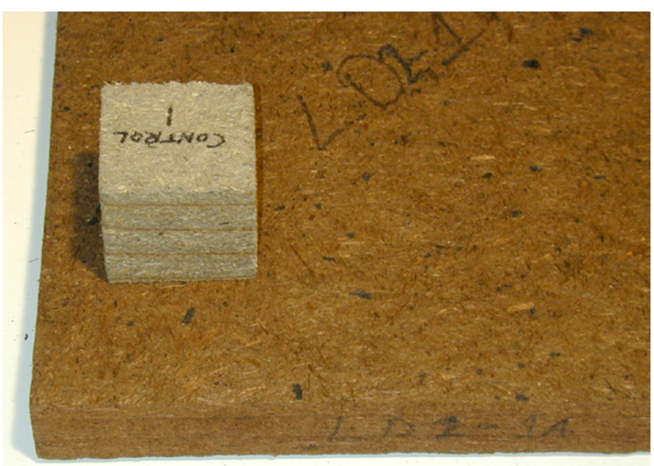


Figure 14. Thermal conductivity sample after conditioning at 121ºC for 21 weeks, compared to a control sample which has seen no elevated

Figure 15. Sample MSC-4 after compression testing, perpendicular orientation. Sample was conditioned at 121ºC for 47 weeks.

temperature.

Each of the various sample types typically contains a few glue joints between fiberboard layers. Layer separation occurred in several samples to varying degrees. The general observation is that the extent to which the layers were initially bonded together can vary significantly, and the more weakly bonded joints tend to separate as the material dries and experiences shrinkage stress. In a few samples, layers completely separated (i.e. the sample separated into 2 sections).

Fiberboard is manufactured primarily as a material for the construction industry, and does not always exhibit the degree of uniformity typically expected for application with nuclear materials. As an example of this, a number of foreign objects have been identified in the fiberboard samples. This includes fragments of rock, glass, aluminum beverage cans and plastic bags. These observed objects have been small and relatively rare, and probably have no significant impact on overall bulk properties.

Conclusions

Additional testing will continue following increasing exposure periods. Ultimately, the data will support development of a predictive model to establish an expected service life for the 9975 packages in KAMS. Several trends and observations are suggested by the data at this time. Specifically:

- The various properties (thermal conductivity, specific heat capacity, compression strength, weight, density) vary at different rates within a given environment. For example, at 121ºC, specific heat capacity shows little change after ~1 year, the compression strength experiences a noticeable decrease around 16 weeks, and the thermal conductivity, weight and density show continual steady decrease over the entire period.
- Where observed, the degradation to date varies both with temperature and humidity. Little change is seen in any measured property at temperatures of 51ºC and below. Changes are generally greater with elevated humidity levels than in dry environments.
- At elevated temperatures, fiberboard weight generally decreases faster than density. This indicates that dimensions are decreasing along with weight, to reduce the effect of weight loss on density.
- The specific heat capacity has not changed significantly in aging studies to date. The total heat capacity of the package is derived from both specific heat capacity and fiberboard weight. Fiberboard weight has shown changes over time in the current data.

References

[1] DOE-STD-3013-2000, "DOE Standard Stabilization, Packaging, and Storage of Plutonium Bearing Materials", September 2000.

[2] "Mechanical Properties of Fiberboard Overpack Materials in the 9975 Shipping Package", W. L. Daugherty and P. R. Vormelker, presented at ASME Pressure Vessels & Piping

Division Conference, July 17-21, 2005, Denver, Co, and published in conference proceedings.

[3] “Thermal Properties of Fiberboard Overpack Materials in the 9975 Shipping Package”, P. R. Vormelker and W. L. Daugherty, presented at ASME Pressure Vessels & Piping Division Conference, July 17-21, 2005, Denver, Co, and published in conference proceedings.