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Aging Model for Cane Fiberboard Overpack in the 9975 Shipping Package

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Abstract

Many radioactive material shipping packages incorporate a cane fiberboard overpack 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. Changes in each of the properties have been fit to a model to allow predictions of degradation under various storage scenarios. Additional data continue to be collected to provide for future refinements to the model.

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 degradation of cane fiberboard after aging, and preliminary models for service life prediction based on those data.

The maximum normal operating temperature in the KAMS storage area is ~50°C. Steady state thermal analysis of the 9975 package predicts this would produce a maximum internal fiberboard temperature of ~85°C. Off-normal or accident conditions can produce higher fiberboard temperatures, up to 121°C. The 9975 shipping package safety analysis allows fiberboard operating temperatures of up to 121°C. Humidity levels inside the drum can vary depending on the initial fiberboard moisture content and storage conditions.

Test Plan

The fiberboard material must retain its dimensions and density within certain ranges to provide the required impact resistance, criticality prevention 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 data of mechanical and thermal properties [2, 3] and test data following environmental exposures of up to 72 weeks [4] have been previously reported. Data following environmental exposures of up to ~200 weeks have now been collected and evaluated statistically to develop preliminary predictive aging models. Conditioning environments include nominally dry environments at 51 to 121ºC, and humidity controlled (30 to 70% relative humidity) environments at 25 to 85ºC.

Test samples were taken from several different packages, and therefore include some degree of fiberboard batch variation.

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).

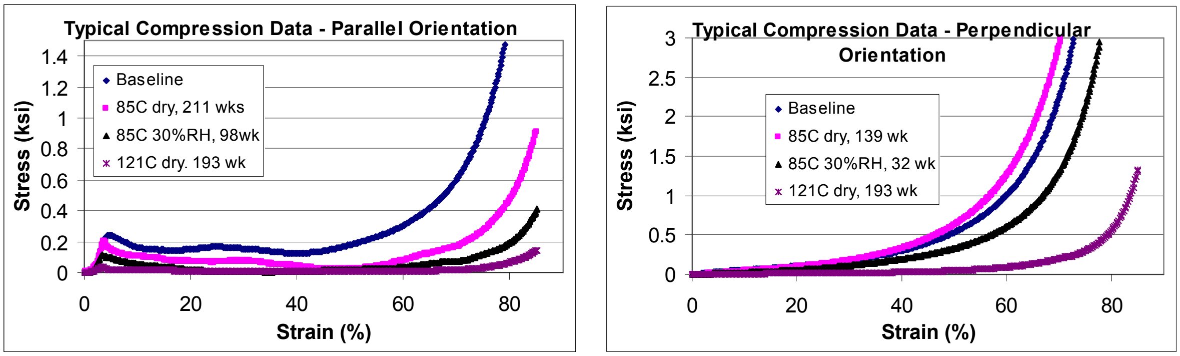
Thermal conductivity samples are typically about 30 x 30 x 3 cm, although some samples are ~18 x 18 x 3 cm. Thermal conductivity is measured on fiberboard samples using a Lasercomp Fox 300 instrument at mean test temperatures of 25, 50 and 85ºC. This instrument provides results consistent with ASTM C518. 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.

Physical measurements (dimensions, weight) are made on samples approximately 5 x 5 x 5 cm. Dimension and weight data have been collected from samples held in each of the aging environments.

Test Data

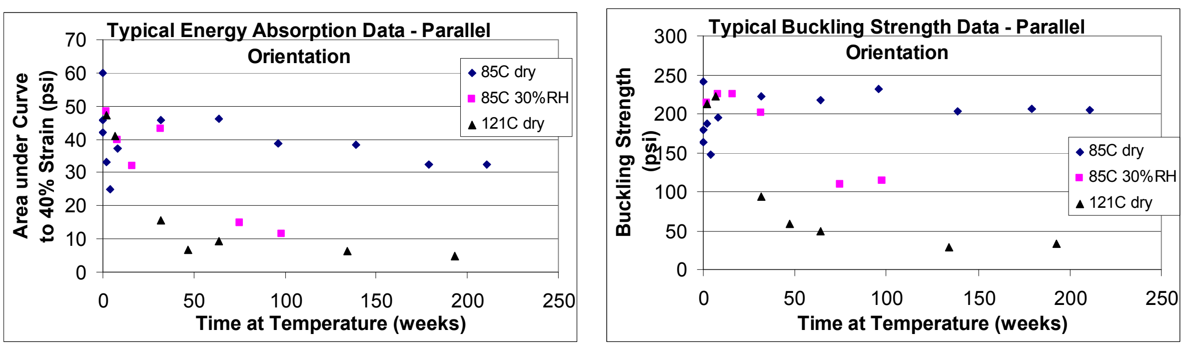
Compression Data

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 package source. Typical compression test stress-strain curves are shown in Figure 1. Typical compression test metrics (area under curve to 40% strain, buckling strength) are shown in Figure 2 for several environments.



(a) Parallel orientation (b) Perpendicular orientation

Figure 1. Typical compression stress-strain curves following environmental exposure, as noted.



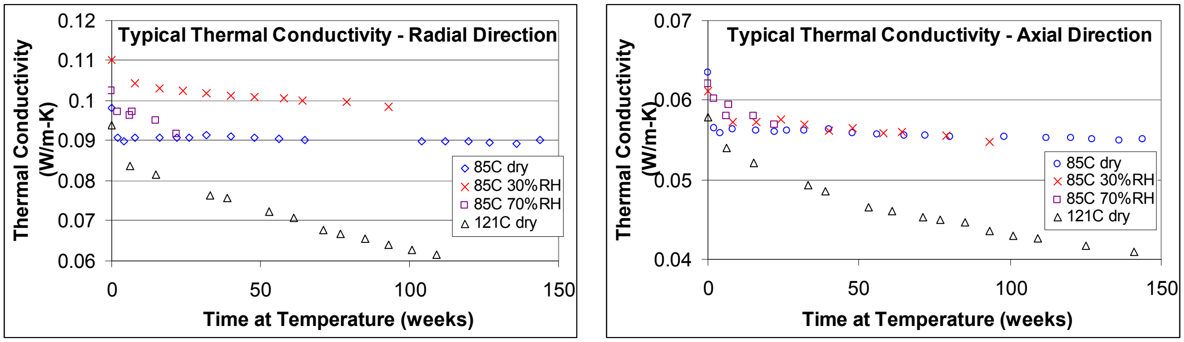
(a) Energy absorption to 40% strain (b) Buckling strength

Figure 2. Typical compression test metrics, parallel orientation, following environmental exposure, as noted.

Thermal Data

Typical thermal conductivity data for a mean test temperature of 25C are summarized in Figure 3. 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 (due to moisture loss). In contrast, increasing loss of thermal conductivity over time is seen as the humidity increases to 30 and 70%RH.



(a) Radial orientation (b) Axial orientation

Figure 3. Typical fiberboard thermal conductivity following conditioning, as noted.

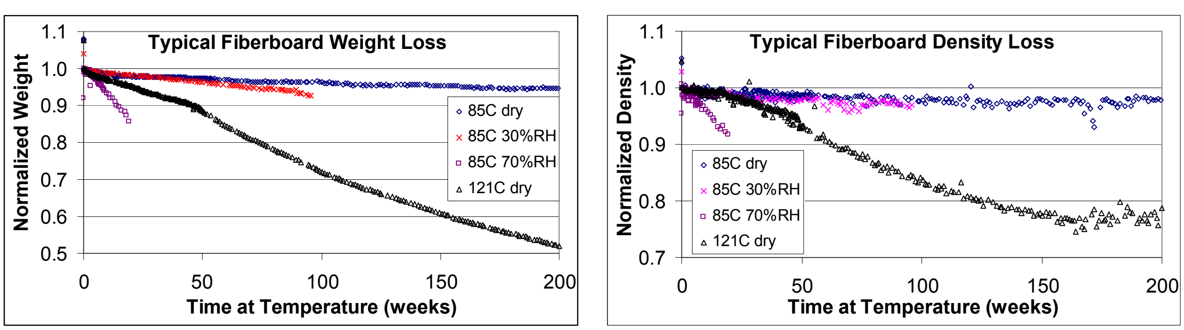
The specific heat capacity was tested on multiple samples following exposures to the same environments as the thermal conductivity samples. These test data show a significant degree of scatter, but no significant change over time. Average specific heat capacity values through 64 weeks exposure in most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

Dimensional and Density Data

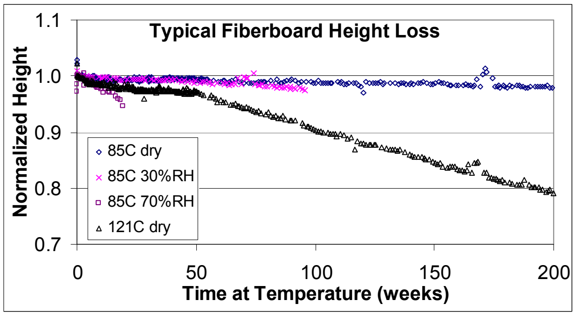
Dimensional and weight information have been collected on several samples in each environment. Since each sample has a different initial weight, the data are compared on the basis of relative weight change. All data are normalized to the sample weight after the first measurement following exposure environment. This eliminates the effect of initial change from moisture loss, and allows multiple samples to be easily compared. Typical data for weight, density and height change in several environments are shown in Figure 4.

Weight loss is observed with time in each environment above 51C. No significant change is observed in the 51C and cooler 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 package sources.

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 as the weight change 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.



(a) Normalized weight change (b) Normalized density change



(c) Normalized height change

Figure 4. Typical fiberboard physical property changes following conditioning, as noted.

Aging Models

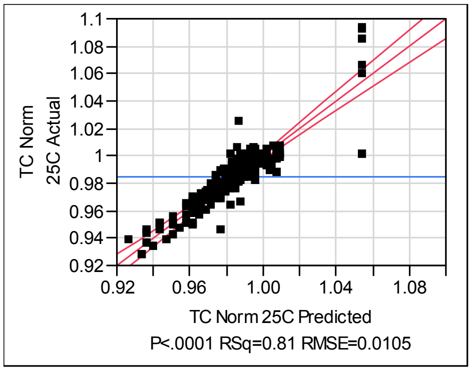
Statistical analysis of the data set for each property has been performed with the goal of developing a predictive model for degradation rates relevant to a range of service environments. Several of these models are described herein to illustrate their development and application.

To improve the modeling accuracy, the data for each property was divided into two regimes. All data at temperatures of 102C and below form the primary regime. This regime includes significant data from dry and elevated humidity environments. The second regime includes all data at temperatures of 102C and above. Since this grouping includes no elevated humidity environments, models for this regime do not capture the effects of humidity. Several models describing the primary regime are described below.

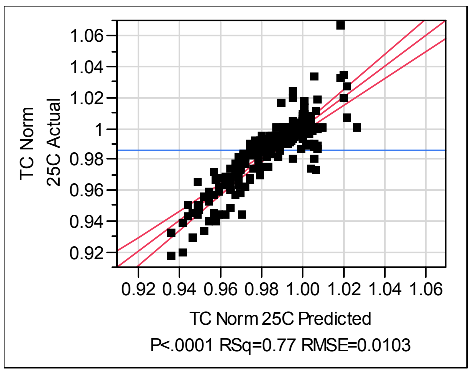
Thermal Conductivity Model

Normalized thermal conductivity was modeled as a function of conditioning time, temperature, relative humidity and the initial density of the sample. Model estimation and the subsequent assessment were conducted using the JMP® [5] statistical software. Initially, a full response surface model (all linear, quadratic and second order interaction terms) was fit to the thermal conductivity data. Separate fits were done for the axial and radial orientation as well as for the different testing temperatures. Terms that were not significant were eliminated from the model and then the model was refit. The process continued until all terms were significant in the model. The resulting fit for the test temperature at 25 deg C (TC25) for the axial and radial orientations are presented in this report (Table 1). Initial density was not significant for predicting TC 25 for either orientation.

The amount of variance explained (R^2) by the TC25 model is about 81% for the axial orientation model (Figure 5a) and about 77% for the radial orientation model (Figure 5b). The relative error [(root mean square error)/mean] for the axial and radial TC25 models is approximately 1%.



(a) Axial orientation



(b) Radial orientation

Figure 5 Actual vs predicted normalized thermal conductivity at 25C

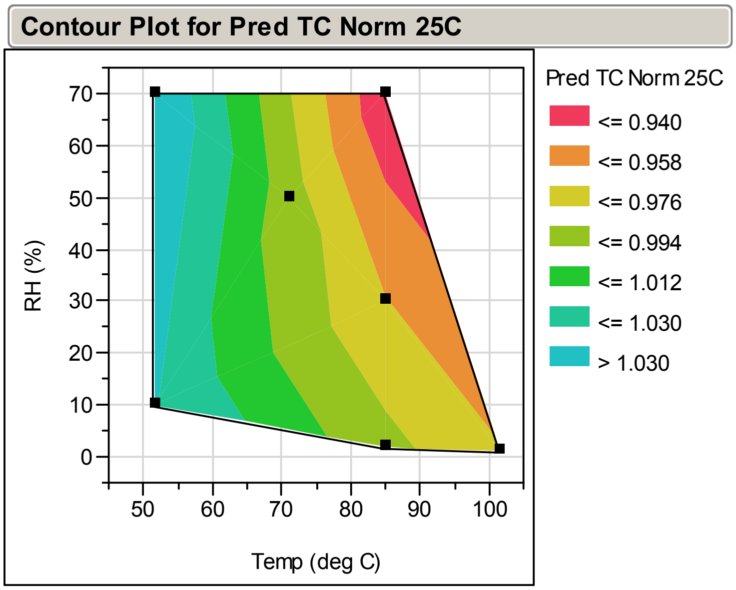
Table 1
Normalized Thermal Conductivity as a function of Conditioning Time, Temperature and Percent Relative Humidity for Conditioning Temperature Range: 51 to 102 deg C *

Term	Axial TC25 Estimate	Std Error	Radial TC25 Estimate	Std Error
Intercept	9.6E-01	8.8E-03	8.8E-01	1.8E-02
Temp (deg C)	4.6E-04	1.0E-04	3.4E-03	4.6E-04
RH (%)	2.8E-03	2.0E-04	1.5E-03	1.9E-04
Time (wk)	1.4E-03	1.5E-04	6.8E-04	7.1E-05
Temp (deg C)*Temp (deg C)	0.0E+00	0.0E+00	-2.2E-05	2.9E-06
Temp (deg C)*RH (%)	-3.4E-05	2.6E-06	-1.9E-05	2.6E-06
Temp (deg C)*Time (wk)	-1.8E-05	1.6E-06	-9.7E-06	8.2E-07
RH (%)*Time (wk)	-7.5E-06	1.7E-06	-1.4E-05	1.7E-06
R^2	81%		77%	
RSME	0.01		0.01	

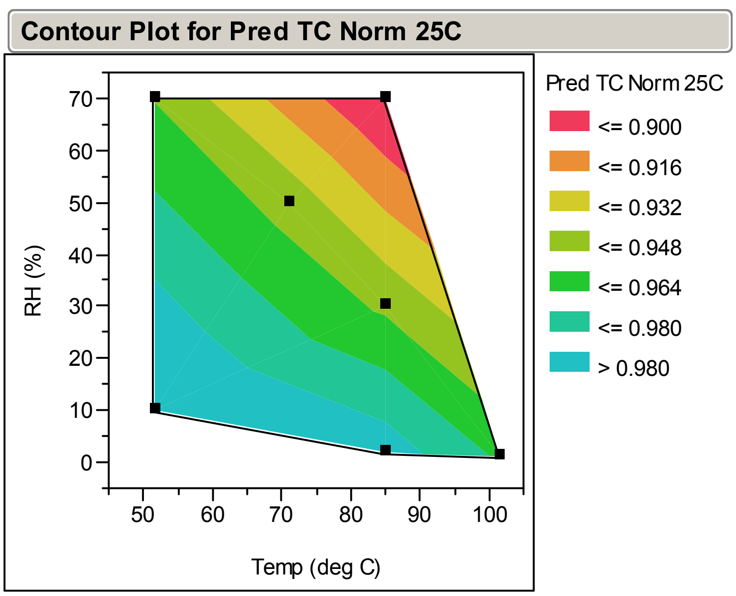
* For example, at 70C, 40 %RH and 2 years (104 wks), the predicted normalized axial thermal conductivity is:

9.6E-1 + (4.6E-4 * 70C) + (2.8E-3 * 40%) + (1.4E-3 * 104 wk) – (3.4E-5 * 70C * 40%) – (1.8E-5 * 70C * 104 wk) – (7.5E-6 * 40% * 104 wk) = 0.99
(or a 1% decrease in axial thermal conductivity).

The axial and radial TC25 models were used to produce the contour plots in Figures 6a and 6b for 100 weeks conditioning time. In addition, the design settings (black dots) are displayed on the contour surface. The greatest reduction in thermal conductivity occurs at the upper right extreme of the factor space (high temperature and high relative humidity) for both the axial and radial orientations. The axial orientation contour plot shows that there is not much impact of humidity for temperatures lower than 80 degrees C whereas the radial contour plot shows a graduated decrease.



(a) Axial orientation



(b) Radial orientation

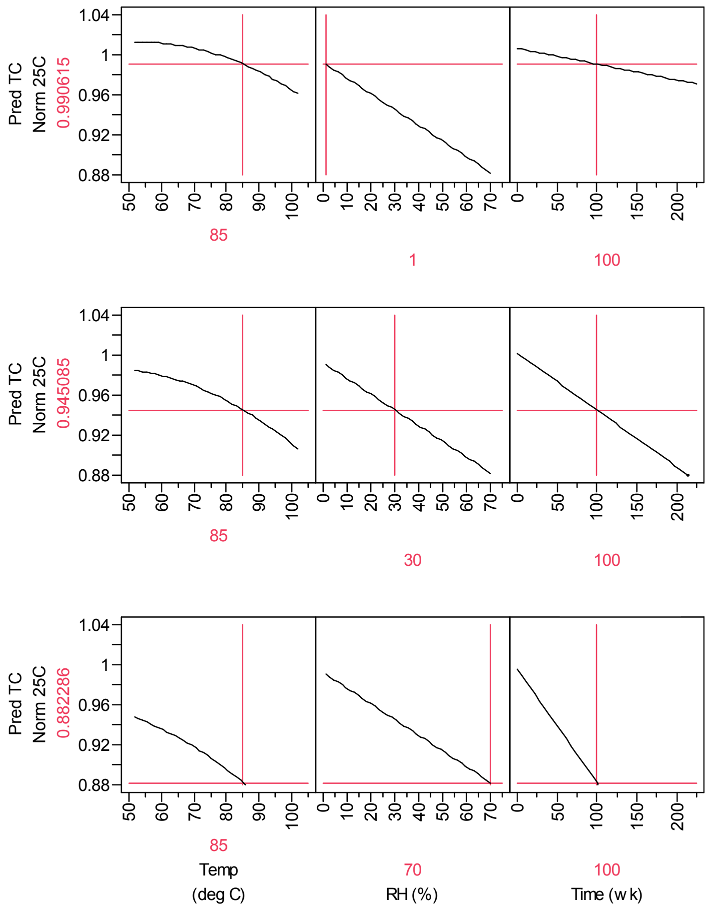
Figure 6 Contour plot for thermal conductivity at 25C for 100 week conditioning time

Prediction profile plots, based on the models in Table 1, were used to assess the impact of conditioning time, temperature and relative humidity and their interactions on thermal conductivity.

Prediction profile plots, available interactively in JMP®, are displays of the predicted response as one design variable is changed while the other variables are held constant. Prediction profiles are especially useful for exploring multiple-response models (e.g.: TC25 for both axial and radial orientations) or as a function of the design variables (Figures 7a – 7c). The low and high values for the process parameters are shown on the horizontal axis. The vertical red dotted line for each process parameter shows its current value or current setting. For each process parameter, the value above the factor name is its current value. The horizontal dotted line shows the current predicted value of each quality measure for the current values of the process parameters. The lines within the plots, called the prediction trace, show how the predicted value changes as the current value of a process parameter is changed. The prediction profile allows the determination of the effect on the predicted response of changing one process parameter at a time. The importance of a factor can be assessed by the steepness of the prediction trace.

If there are interaction effects (cross-product effects in a model), the prediction traces can shift their slope and curvature as the current values of the process parameters are changed. If there are no interaction effects, the traces only change in height, not slope.

The profile plots show the impact of conditioning time for the radial orientation. A similar impact is seen for the axial orientation. Thermal conductivity decreases as conditioning time, relative humidity or temperature increase.
Figures 7a-7c (conditioning temperature fixed at 85 degrees C) show that the importance of conditioning time increases as relative humidity increases from 1% to 30% and finally to 70%.



Prediction Profile for Radial TC25
Temp= 85 deg C, RH=1%, 30% and 70% and Time=100 weeks
Figure 7

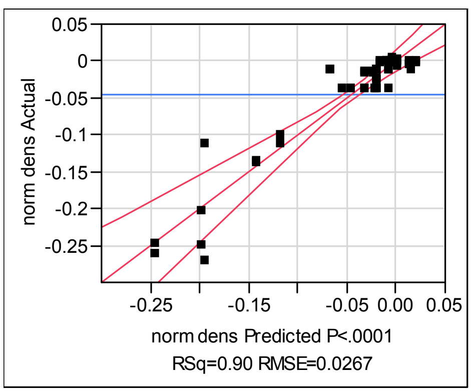
Density, Weight and Sample Height Models

The impact of conditioning time, humidity, temperature and material package source on sample density, weight and height were modeled. The data after the start of conditioning were visually screened for outliers resulting in a small amount of data being eliminated. The first modeling attempt was similar to that for thermal conductivity. Specifically, the conditioning variables and material source was used to predict the normalized metrics (normalized density, weight and height). Adequate model fits did not result primarily due to different degradation rates for repeat samples under the same conditioning.

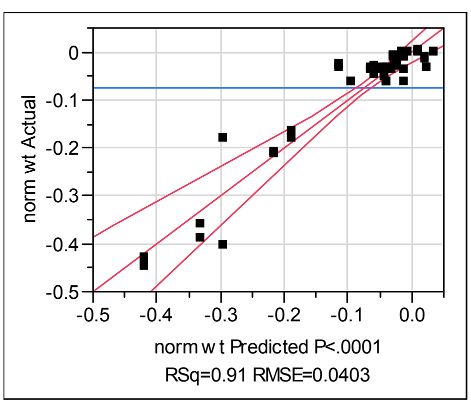
The normalized metrics for density, height, and weight were plotted versus time (e.g. Figure 4). Visual review of the plots suggested that each sample could be reasonably modeled by a straight line time trend. The slope estimates for the change in each metric over time, a measure of material degradation, were then used as the dependent variable related to the conditioning factors of temperature, humidity and sample material source. The slopes that were not significant were set to zero before conducting the statistical modeling.

The resulting models for the slope (change in the normalized metric versus time) of normalized density, weight and height are displayed in Table 2. The material package sources coded as 2234, LD1, LD2, MSC and New were considered as categorical variables in the modeling. The estimates for coefficients of these variables, displayed in Table 2, are for the listed material source relative to "New" package source material. The interaction of temperature and relative humidity with the 2234 package source material was significant while many of the remaining interactions with material package source were not significant. However, these terms were left in the models because of the improvement observed in reviewing the various actual versus predicted plots (Figures 8a-8c).

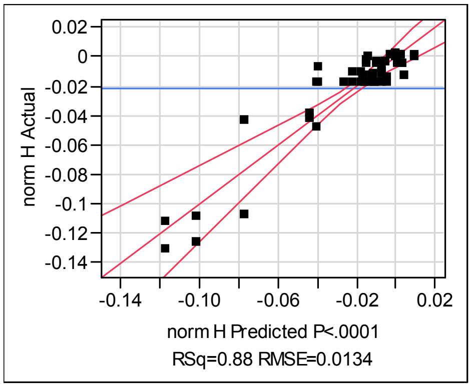
The conditioning factors are all significant and their estimated coefficients maintain the same sign across all models. The material package source 2234 had a significant impact on interaction terms for both conditioning temperature and relative humidity. The quality of fit (perfect being 100%) for each model is indicated by the R^2 and is greater than 88% for the models in Table 2. The predictive ability is reasonable for the mean slope of all three models. However, the uncertainty as exhibited in the actual versus predicted plots (Figures 8a, 8b and 8c) is appreciable and reflects degradation differences among the samples that were conditioned.



(a) Normalized Density



(b) Normalized Weight



(c) Normalized Height

Normalized Height
Figure 8 Actual vs. predicted for slope of indicated property

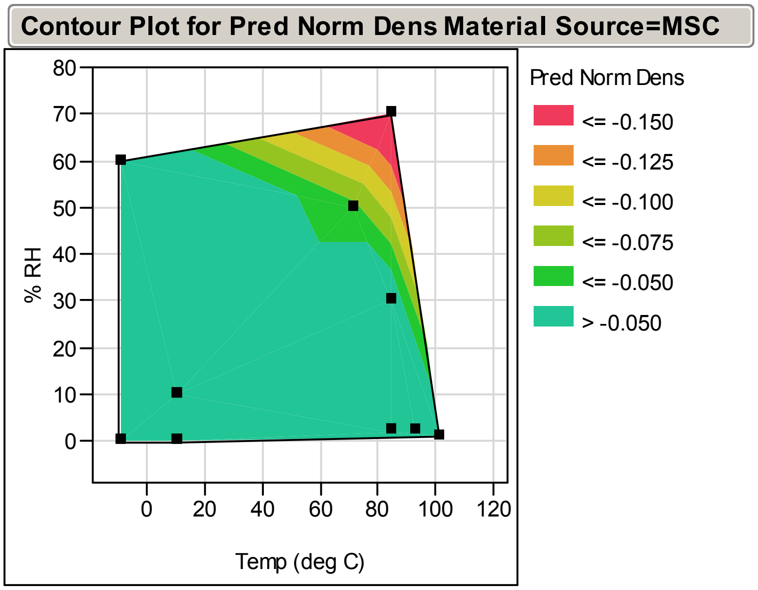
Table 2
Model for Slope of Normalized Density, Weight and Height as a function of Conditioning Temperature, Percent Relative Humidity and Material Package Source for Conditioning Temperature Range: -10 to 102 deg C *

Term	Normalized Density Estimate	Standard Error	Normalized Weight Estimate	Standard Error	Normalized Height Estimate	Standard Error
Model values for material package source New						
Intercept	3.6E-02	4.9E-02	1.1E-01	7.4E-02	2.0E-02	2.4E-02
% RH	3.9E-03	7.4E-04	6.5E-03	1.1E-03	1.9E-03	3.7E-04
Temp (deg C)	-5.5E-04	5.1E-04	-1.5E-03	7.8E-04	-3.5E-04	2.6E-04
% RH*% RH	-6.6E-05	9.6E-06	-1.1E-04	1.5E-05	-2.7E-05	4.8E-06
% RH*Temp (deg C)	-2.1E-05	4.9E-06	-3.5E-05	7.4E-06	-1.1E-05	2.5E-06
Adjustments to model values for alternate material package sources						
[2234]	2.0E-01	8.7E-02	3.3E-01	1.3E-01	1.1E-01	4.4E-02
[LD1]	5.0E-02	8.7E-02	5.4E-02	1.3E-01	2.6E-02	4.4E-02
[LD2]	-8.9E-02	8.7E-02	-8.2E-02	1.3E-01	-2.8E-02	4.4E-02
[MSC]	-4.4E-02	4.8E-02	-1.2E-01	7.3E-02	-2.2E-02	2.4E-02
% RH*[2234]	-1.3E-03	3.4E-04	-2.4E-03	5.2E-04	-7.6E-04	1.7E-04
% RH*[LD1]	-2.6E-04	3.4E-04	-1.1E-04	5.2E-04	-4.9E-05	1.7E-04
% RH*[LD2]	5.2E-04	3.4E-04	1.1E-03	5.2E-04	5.1E-04	1.7E-04
% RH*[MSC]	-7.2E-05	3.0E-04	-3.0E-04	4.6E-04	-3.4E-04	1.5E-04
Temp (deg C)*[2234]	-2.1E-03	9.4E-04	-3.4E-03	1.4E-03	-1.2E-03	4.7E-04
Temp (deg C)*[LD1]	-5.5E-04	9.4E-04	-6.0E-04	1.4E-03	-2.8E-04	4.7E-04
Temp (deg C)*[LD2]	1.1E-03	9.4E-04	9.2E-04	1.4E-03	2.9E-04	4.7E-04
Temp (deg C)*[MSC]	3.5E-04	5.3E-04	1.1E-03	8.0E-04	2.3E-04	2.7E-04
R^2	90%		91%		88%	
RMSE	0.027		0.040		0.013	

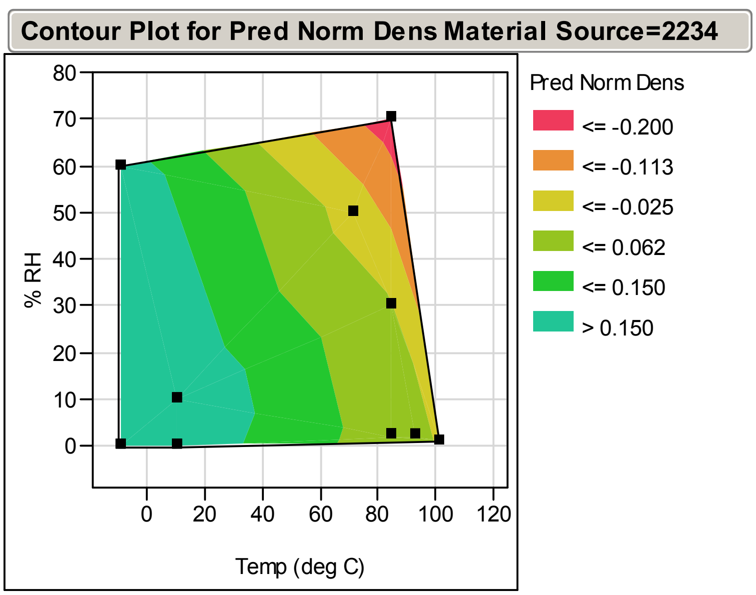
* For example, at 70C and 40 %RH, the predicted normalized rate of change in density for MSC material is:

3.6E-2 + (3.9E-3 * 40%) - (5.5E-4 * 70C) - (6.6E-5 * 40% * 40%) – (2.1E-5 * 40% * 70C) – 4.4E-2 - (7.2E-5 * 40%) + (3.5E-4 * 70C) = -0.03
(or a 3% decrease in density per year).

The models in Table 2 were used to produce the contour plots of normalized density in Figures 9a for MSC material and 9b for 2234 material. The design settings (black dots) are also displayed on the contour surface. The models can be used for interpolation within the design space but should not be used for extrapolation. The steepest predicted slope occurs at the upper right extreme of the factor space (high temperature and high relative humidity) for the slope of normalized density. Predicted normalized density covers a greater range over the factor space for 2234 than for the MSC material package source. Similar behavior was observed for normalized weight and height.



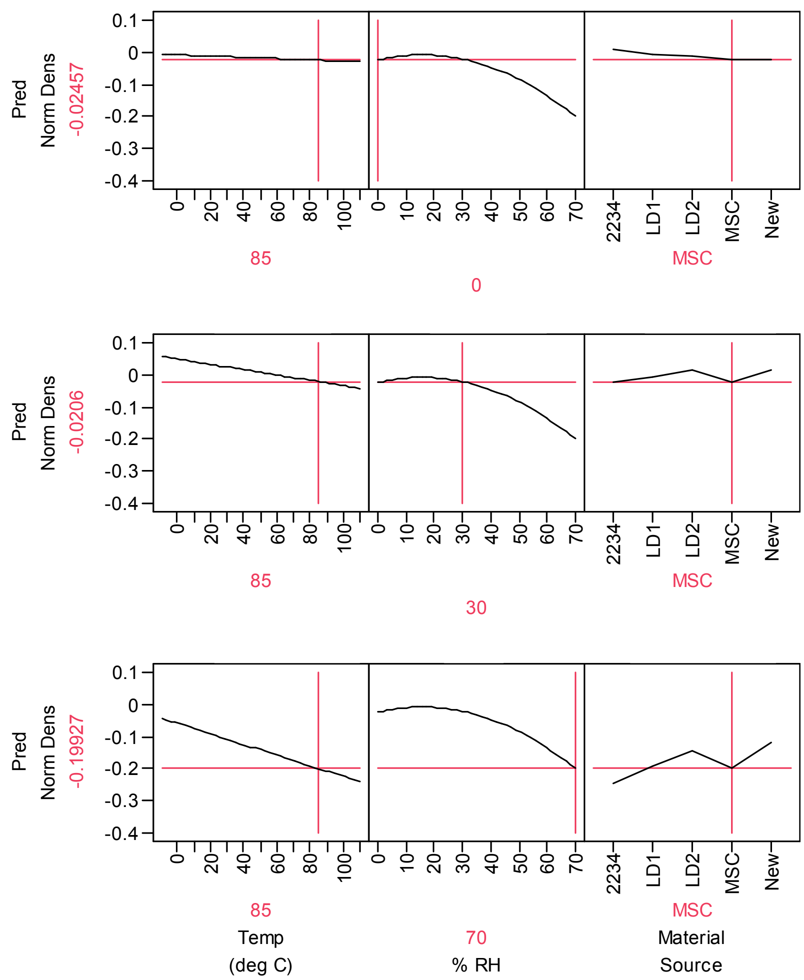
(a) Material Package Source MSC



(b) Material Package Source 2234

Figure 9 Contour Plot for Slope (change in Normalized Density per Year)
of Normalized Density (For example, a predicted change in normalized density slope of -0.1 corresponds to a 10% decrease in actual density per year.

The profile plots show the impact of conditioning temperature and relative humidity on the slope of predicted normalized density for the MSC material package source. The slope decreases as temperature and humidity increase. At dry conditions (Figure 10) the impact of temperature is minor but increases in importance as humidity increases.



Prediction Profile for Slopes of Normalized Density,
Temp= 85 deg C, RH=0%, 30% and 70%, Material Package Source=MSC
Figure 10

<u>Conclusions</u>

Additional testing will continue following increasing exposure periods. A predictive model has been developed for each property, which provides an estimate of service life for the package under storage conditions. Refinements to each model will continue to be made as additional data is collected. Several trends and observations are suggested by the data at this time. Specifically:

- Degradation of each property (thermal conductivity, specific heat capacity, compression strength, weight, density) occurs at different rates within a given environment. Therefore, separate aging models are required for each property.
- 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.

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.

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[5] JMP® Version 7.0, SAS Institute, Cary, N.C.